Broadly tunable parametric line emission from $\beta$-barium borate on pumping with picosecond pulses

Ambika Nautiyal, Prem B. Bisht *

Department of Physics, Indian Institute of Technology Madras, Chennai 600 036, India

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Abstract

Tunable line emission (LE) over a large wavelength region (340–980 nm) is obtained by pumping thick crystals of $\beta$-barium borate (BBO) with picosecond pulses at 532 nm. Phenomena of group velocity dispersion, diffraction and phase matching take place simultaneously such that the radiation shows specific features that are characteristic of (i) strongly coupled fundamental and harmonic fields, (ii) amplified phase matched superfluorescence, (iii) conical emission and (iv) four wave mixing.

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1. Introduction

Tunable sources of short duration pulses are required to probe the ultrafast processes in physics, chemistry, and biology [1]. Noncollinear optical parametric amplification (NOPA) process provides such a stable source [2,3]. The attention has also shifted to chirped pulse optical parametric amplifiers (CPOPA) in recent years due to the need of high power short pulses. The requirement of seed pulses for these applications remains a subject of investigations from the points of view of (i) tunability, (ii) stability and (iii) their suitability for existing pump sources. At present, the phenomena popular for such applications are white light continuum, cw sources and optical parametric generation (OPG) [4,5].

The interplay among Kerr nonlinearity, group velocity dispersion (GVD) and diffraction has been reported to induce the phenomenon of modulation instability (MI) in nonlinear optical crystals [6,7]. In a nonlinear medium, the dynamic interaction of strongly coupled fundamental and harmonic fields has been shown to induce the breakup of input beams into a line of circularly separated beams known as conical emission [8]. Under femtosecond (fs) pumping of 6 mm thick crystal of $\beta$-barium borate (BBO), a tunable emission has been observed from 480 nm to 800 nm [9].

Recent experimental observations on self-focusing of intense ultrashort pulses indicate that spatial and temporal degrees of freedom are to be treated together [10]. Spatio-temporal MIs in quadratic bulk samples were predicted to cause exponential growth of perturbation, which results in the line emission. Optical parametric spectral broadening of picosecond pulses has been reported in literature on pumping with 355 nm and 266 nm [11]. During the trial experiments on noncollinear optical parametric amplification of white light continuum under picosecond (ps) pumping at 532 nm [12], we came across a tunable radiation from 340 nm to 980 nm. Considering the extraordinary broad tuning range in near UV, visible and near IR regions, we report its spectral characteristics in detail here.
2. Experimental

Fig. 1 shows the schematic of the experimental setup. Picosecond pulses from a Nd:YAG laser at 1064 nm (10 Hz, 35 ps) were frequency doubled (532 nm) with pulse energies of 2 mJ. The beam was focused by a concave mirror of focal length 500 mm on a BBO crystal (Type I, 21.9°). The calculated spot size of the beam on the front surface of the crystal was 0.6 mm. An iris diaphragm and the neutral density filters were used to change the incident fluence on the crystal. The pump beam (s-polarized) was sent perpendicular to the BBO surface. The BBO crystal was tilted further to achieve the angle-tuned phase matching for various frequencies. Spectra of generated radiations were recorded with spectrometer (Ocean Optics, HR 2000) with a resolution of ~5 nm.

3. Results and discussion

Fig. 2 shows the photograph of the obtained (visible) colours taken from the screen at different tilt angles for a 12 mm thick BBO crystal. It can be seen that for a tilt of ~1.5°, a yellowish-red spot is observed above the pump beam. The spot shifts away from the pump beam on increasing the tilt of the BBO crystal. At a large tilt angle (~12°), many spots are observed distributed about the pump beam. The regions of the invisible colours lie in the near IR range (750–980 nm). At smaller tilt angles, the angular positions of the observed colours were found to be the function of their frequency. The propagation angle (θ) with respect to the input beam can be given by the relation [13]

$$\phi = \sqrt{\frac{k_0^2}{k_0^2 - k^2}} \Omega,$$

where $k_0 = 2\pi n/\lambda_0$, ($\lambda_0$ is the pump wavelength and the $n$ is the refractive index of the optical medium), $k_0^2$ is the group velocity dispersion given by $\frac{d^2 k}{d\omega^2}$ and $\Omega = \omega - \omega_0$ is the difference between the generated frequency and the input frequency.

3.1. Observation of conical type emission at low tilt angles

3.1.1. At a fix angular position of the detector

Fig. 3 gives the spectra of the LE obtained at a typical position of the detector (φ = 2°) as a function of the crystal tilt angle (θ). Here, spectra are continuously tunable between 530 nm and 700 nm. On increasing the crystal tilt, the spectra shift towards the higher energy. It was also observed that the full width at half maximum (FWHM) of the spectra decrease with increasing the tilt. The decrease in the spectral bandwidth ($\Delta \lambda$) is given by Eq. (2), which relates it with the effective length of crystal ($\ell_2$) and the peak wavelength ($\lambda$) as [14]

$$\Delta \lambda = \frac{\lambda^2}{c} \frac{|u_{ai}|}{\ell_2},$$

where $c$ is the speed of light and

$$1 = \frac{1}{u_{ai}} = \frac{1}{v_i \cos(\alpha + \beta)} - \frac{1}{v_i}.$$

Here $v_i$ and $v_s$ are the group velocities of idler and signal waves, $\alpha$ and $\beta$ are the angles made by the pump with the signal, and the idler waves, respectively. As seen from Fig. 1, the pump is collinear and the signal angle inside the crystal is smaller than φ for the range of wavelengths obtained in Fig. 3. Therefore, considering the nearly collinear geometry for the observed peak values as signal wavelengths, the ratio of the values of $\Delta \lambda$ calculated from Eq. (2) matches well with those obtained experimentally. The broad spectrum at 1.5° tilt, can also be fitted with a sum of two Gaussian curves indicating the possibility of overlap of two different emission peaks.

3.1.2. At a fixed tilt angle of the crystal

Table 1 gives the observed angular positions (φ) of the radiations for crystal tilt (θ) fixed at 6°. The calculated values of the angular positions (φ) of the emissions (by using Eq. (1)) are also given for the comparison. It is seen that the calculated values of φ match reasonably well with those obtained experimentally. At large tilt angles however, due to the internal reflection within the long (12 mm) crystal, the values of φ are arbitrary and hence do not follow Eq. (1).

3.2. Behaviour at large tilt angles

Spectra of the emissions recorded by vertically translating the detector (varying φ) are given in Fig. 4. Here the tilt angles of the BBO crystal are in the range of 10–16° from the vertical axis. Table 2 gives the peak positions and the observed shifts from the pump beam (532 nm). It can be seen that the FWHM of some of the spectra are broad. The broadening may be due to the overlap of the various emissions, or group velocity dispersion effects.
A BBO crystal cut an angle of 21.9° is suitable for the phase matching in the wavelengths near 625–650 nm regions at the pump wavelength of 532 nm [15]. By further tilting the crystal the phase matching wavelength range can be increased. Calculations were done by using the standard software [16]. The reliability of the program was checked by reproducing the curves published in the literature [17]. The experimentally observed peak positions were obtained from simulations by adding the given tilt to the crystal.

3.3. Analysis of the observed line emissions

On the basis of the above discussion, the observed peaks listed in Table 2 can be assigned prominently with two

### Table 1

<table>
<thead>
<tr>
<th>Obtained wavelength (nm) ±2 nm</th>
<th>Observed angular positions (°) ±0.5°</th>
<th>Calculated angular positions (°) ±0.3°</th>
</tr>
</thead>
<tbody>
<tr>
<td>941</td>
<td>6.8</td>
<td>7.0</td>
</tr>
<tr>
<td>844</td>
<td>5.8</td>
<td>5.6</td>
</tr>
<tr>
<td>655</td>
<td>2.5</td>
<td>3.0</td>
</tr>
<tr>
<td>550</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>532 (pump)</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>424</td>
<td>–4.3°</td>
<td>–3.6°</td>
</tr>
</tbody>
</table>

Negative sign indicates that the radiation was observed below the pump spot.

A BBO crystal cut an angle of 21.9° is suitable for the phase matching in the wavelengths near 625–650 nm regions at the pump wavelength of 532 nm [15]. By further tilting the crystal the phase matching wavelength range can be increased. Calculations were done by using the standard software [16]. The reliability of the program was checked by reproducing the curves published in the literature [17]. The experimentally observed peak positions were obtained from simulations by adding the given tilt to the crystal.

3.3. Analysis of the observed line emissions

On the basis of the above discussion, the observed peaks listed in Table 2 can be assigned prominently with two
mechanisms. Firstly, most of the radiations observed in the region of 625–980 nm \((v_2–v_{10})\) can be assigned to the amplification of the superfluorescence at the pump wavelength of 532 nm. The idler frequencies of these signals lie beyond the detection limits (<1100 nm) of the present system. Secondly, some lines \((v_5, v_6)\) can be assigned as the idler frequencies along with the corresponding signal frequencies \((v_{14}, v_{13})\) in the 400–500 nm range generated through the two-photon induced four wave mixing (FWM). A few of the observed radiations (e.g., \(v_5, v_6\)) can be assigned by both the mechanisms. In addition, some signals are due to the SHG \((v_{12}, v_{13})\) and the SFG \((v_{16}, v_{17})\) of the existing signals as indicated in the Table 2. Even after the use of dichroic mirrors, there is a small contribution \(<1\%\) of the fundamental wavelength (1064 nm) in the system. As a result, a sum frequency generation may occur between the fundamental and second harmonic of the laser. Therefore, 355 nm (also observed in the experiment) may be considered as the pump wavelength for amplification of various emissions. Further, as indicated by the curve 10, besides the line emissions with sharp peaks, we also observed broad emissions containing a few peaks at the tilt angles of \(8–10^\circ\). These may indicate towards the white light continuum type radiations reported earlier by Smith et al. [18] and Yang and Shen [19].

### 3.4. Observation of retracing behaviour

Fig. 5 gives the theoretical simulations for phase matching angles for amplified signal wavelengths at the pump wavelength of 532 nm. The inset shows the experimentally observed spectrum with at a small crystal tilt of 0.8\(^\circ\). It is seen that the theoretically simulated curve exhibits the phenomenon of retracing behaviour. Pairs of the signal wave-
a small tilt provides additional phase matching for an additional pair of wavelengths.

3.5. Effect of thickness of the crystals

Experiments were also performed by using a BBO crystal of thickness 20 mm. In this case, the tunability was limited as compared to that observed with the 12 mm thick crystal. This is due to the restriction of light passage through the thicker crystal at large tilt angles. Further, it is interesting to note that at the same pump energy, no emission could be observed from a 2 mm thick crystal. This indicates towards the fact that the generation of the superfluorescence in the first few mm of the thick crystal helps in observation of amplified radiations in the present case. This is consistent with the reported observations in BBO [21], where the cascaded second harmonic generation (SHG) and subsequent sum frequency generation (SFG) processes have been found to contribute significantly even at non-phasematched angles.

4. Conclusions

In conclusion, to the best of our knowledge, for the first time, we have observed broadly tunable radiations from thick BBO crystals on pumping with second harmonic of a ps Nd:YAG laser. The observed radiations have been assigned on the basis of optical parametric amplification and two photon induced FWM by pumping with second harmonic of the Nd:YAG laser. The results obtained here indicate the various phase matching conditions fulfilled due to ps laser pulses in long crystals with varying large tilts. It is due to the fact that only long crystals have the capability of generating other radiations (in first few mm of their length) that act as the sources for a particular new radiation when the phase matching condition is satisfied. This is facilitated by various larger tilts. It is believed that radiations so obtained may be useful as seeds for tunable ultra-short pulse generation using the NOPA processes and pulse compression techniques.

Acknowledgement

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References