Colored conical emission in BBO crystal induced by intense femtosecond pulses

Jie Bi\textsuperscript{a,b}, Xing Liu\textsuperscript{a}, Yuhua Li\textsuperscript{a,⁎}, Peixiang Lu\textsuperscript{a}

\textsuperscript{a} Wuhan National Laboratory for Optoelectronics, School of OptoElectronic Science and Engineering, Huazhong University of Science and Technology, Wuhan 430074, China
\textsuperscript{b} School of Mathematics and Physics, China University of Geosciences, Wuhan 430074, China

\textsuperscript{⁎} Corresponding author.
E-mail address: lsklab@mail.hust.edu.cn (Y. Li).

1. Introduction

CCE is an attractive nonlinear phenomenon, a large number of experiments have been performed to observe CCE in dense metal vapors [1,2] and ultrashort pulse filamentation in air [3–5] or Kerr media [6,7]. During intense laser pulses propagate in a nonlinear medium, temporal and spatial MI appear due to the interplay of optical Kerr nonlinearity and group velocity dispersion or diffraction respectively, which cause temporal and spatial perturbations with infinite extent in time as temporal breakup [8] or in space as filamentation [9,10]. While as to quadratic bulk samples, MIs occur during the second harmonic generation due to the parametric instability [11–14], the strong coupling of the fundamental and harmonic fields was observed to produce quadratic spatial solitons [11]. Spatiotemporal MIs were predicted theoretically [14,15] and demonstrated experimentally [16,17] to lead to CCE with rings of different colors growing exponentially. CCE can be induced in a quadric bulk medium such as BBO crystal not only by intense fundamental femtosecond (fs) pulses within the process of parametric downconversion and sum-frequency, which conforms to the experimental conical angles of CCE and explains the influence of incident angle on the conical angle. Furthermore, the beam angular spectra have been modified within the process of parametric downconversion and sum-frequency, which conforms to the experimental conical angles of CCE and explains the influence of incident angle on the conical angle.

2. Experimental setup and results

A 1 kHz regeneratively amplified Ti: sapphire femtosecond laser (Spitfire, Spectra-Physics) was applied to produce 50 fs pulses with the maximum pulse energy of about 600 μJ at 800 nm. The experimental setup is shown in Fig. 1(a). A half-wave plate combined with a polarizing plate was employed to adjust the intensity of the laser pulse. An iris was inserted between the laser and the half-wave plate to adjust the beam diameter and change the influence of the fundamental pulses on the generation of CCE accordingly. The laser beam was focused into a 5 mm-thick β-BBO crystal (type-I, 29.18°cut), which was placed carefully near the focus, by a lens of 20-mm focal length. With an appropriate pump power and exactly normal incidence to the BBO crystal, a blue-green cone with a conical angle of about 4.8° appeared evidently around the beam propagation axis, of which the angular width is about 0.5° (4.5°–5.0°), as shown in Fig. 1(b). A spectrometer was used to monitor the spectrum of the blue-green CE, and the peak is approximately at 485 nm, as shown in Fig. 1(c).

As the pump beam was sent perpendicular to the BBO crystal, the angle between the crystal optical axis and pump beam was just 29.18° and it satisfied the phase matching for SHG. Here the BBO crystal was rotated to reduce second harmonic conversion, and the blue-green CE
became weak accordingly. Fig. 2(a) shows the dependence of the blue-green CE energy on the rotation angle of crystal. This clearly indicates that the blue-green CE is originated from coupling between the fundamental and second harmonic pulses. The generation of CCE is critical on the intensity of the incident fundamental pulse and CCE appears only when the intensity is beyond the threshold. With the increase of the incident fundamental energy, the blue-green CE has an obvious gain and becomes much brighter, as shown in Fig. 2(b). The spectrum of the blue-green CE was recorded under different pump intensities, which could be achieved by tuning a half-wave plate combined with a polarizing plate or scanning the BBO crystal straightly along the propagation axis, as shown in Fig. 3(a) and (b), respectively.

![Fig. 1](image1.png)

Fig. 1. (a) Setup employed in the experiment. I, iris with variable diameter; HW, a half-wave plate in 800 nm; P, a polarizing plate; L, lens with focus length of 20 cm; C, 5 mm-thick BBO crystal (type-I, 29.18° cut); (b) A typical picture and (c) the spectra of the generated blue-green CE.

![Fig. 2](image2.png)

Fig. 2. The dependence of blue-green CE energy on (a) the incident angle of pumped pulses to the BBO crystal, (b) the fundamental intensity.

![Fig. 3](image3.png)

Fig. 3. The spectrum of blue-green CE pumped by (a) fundamental pulses (50 fs) after a half-wave plate and a polarizing plate to get variable energy at 9.7 mJ/cm² (solid line), 8.24 mJ/cm² (dotted line), 4.85 mJ/cm² (dashed line), and (b) slightly focused fundamental pulses (50 fs) with the BBO crystal being scanning to get variable intensities at 6.3 mJ/cm² (solid line), 5.0 mJ/cm² (dotted line), 4.1 mJ/cm² (dashed line).
When the BBO crystal was moved closer to the focus, the blue-green CE also became much more intense and brighter. According to the spectra of the blue-green CE shown in Fig. 3, one can see clearly that a slight red-shift appears as the incident intensity decreases by reducing the fundamental energy or moving the crystal away from the focus. While the fundamental energy was increased to about 0.2 mJ, an incomplete yellow circle appeared accidentally outward the blue-green circle with a conical angle of about 6.0°. A typical image and the corresponding spectrum of the yellow CE are given in Fig. 4. In the experimental setup, an iris was placed to vary the incident pulse beam diameter and hence the influence of the incident fundamental pulse. The blue-green CE had the highest intensity when the diameter of the iris was about 2.5 mm, and almost disappeared when the diameter was reduced to less than 1.5 mm or enlarged to more than 8 mm. Different spectra of blue-green CE and yellow CE were recorded in Fig. 5 under changing pump pulse width from 50 fs to 120 fs, which reveals a clear red-shift when the pulse width was stretched. As the FWHM duration of fundamental pulses was negatively chirped from 50 fs by increasing the distance of the grating pair of the CPA system, both the yellow and the blue-green CE became weak, and the yellow CE even completely disappeared at about 120 fs. All these phenomena distinctly revealed that the CCEs were originated from strong spatiotemporal coupling between the fundamental and second harmonica pulses [8,9,15].

In quadratic media the underlying physics of CCE by means of SHG involve multiple three-photon processes [15]. Within the process of phase-matching SHG, second harmonic photons 2ω appear and travel...
on-axis. Then the parametric downconversion process takes places, second harmonic photons $2 \omega$ decay into photon pairs at different frequencies $\omega \pm \delta \omega$ traveling with opposite off-axis angles, which then mix with the on-axis pump fundamental photons at $\omega$ by means of sum-frequency and produce photon pairs at $2 \omega \pm \delta \omega$ that travel off-axis. In our previous work Ref. [17] where CCE pumped by SH pulses was studied, the influences of GVM and GVD on CCE were explained. Similarly the affects of GVM and GVD can be implemented into the CCE generation pumped by fundamental pulses. According to the calculated GVM between on-axis SH and off-axis CCE, as shown in Fig. 6(a), there is minimum GVM locates at 475 nm. While the strongest blue-green CE occurred at 485 nm, and the weak yellow CE occurred at 575 nm, where the corresponding GVM are about 11.6 fs/mm and 95.8 fs/mm respectively. So we can see that CCE withstands the presence of GVM, while CCE with lower GVM appears much more brightly. Here $k_2 = d^2 k/\delta \omega^2$ are GVDs, and $\beta = k''(2 \omega - \delta \omega)/k''(2 \omega)$. As shown in Fig. 6(b), the GVD of CE pulse equal to that of SH at 517 nm. It indicates that both GVM and GVD reach balance between blue-green CE and SH, so frequencies around 485 nm accumulate the greatest gain when propagating in BBO crystal within all different frequencies and generate the strongest cone.

The process of parametric downconversion satisfies the noncollinear phase-matching condition $k(2\omega) = k(\omega + \delta \omega) + k(\omega - \delta \omega)$, the off-axis angles of photon pairs at different frequencies $\omega \pm \delta \omega$ are given by $\sin^2 \theta_{\pm} = k_2(k(\omega \pm \delta \omega))$ and $k_2^2 = k^2(\omega + \delta \omega) - k^2(\omega - \delta \omega)$. The process of sum-frequency also is in the limit of the noncollinear phase-matching $k_2(\omega \pm \delta \omega) + k(\omega) = k(2 \omega \pm \delta \omega)$, the conical angles at different wavelengths are given by $\tan \theta_{\pm} = \sin \theta_{\perp} / [k(\omega \pm \delta \omega) \cos \theta_{\perp} + k(\omega)]$, as shown in Fig. 7. The solid curve indicates that the conical angle sequence of different wavelengths in normal incidence, it is clear that the calculated conical angles corresponding to 485 nm and 575 nm are 43° and 57°, respectively. These calculated results explain the radius of the incomplete yellow ring is larger than that of the blue-green ring, which are in agreement with our experimental observation. Fig. 7 indicates that the entire wavelengths shift to larger conical angles with the incidence angle changing from $-1^\circ$ (tilting down) to $2^\circ$ (tilting up). To confirm this prediction the BBO crystal was rotated in the plane consisting of the optical axis of BBO crystal and the beam propagation axis. CCEs culminated to the highest intensity in normal incidence and decayed gradually as the incident angle increased. Meanwhile, the blue-green circle became larger while the BBO crystal was rotated along counter-clockwise, which is in good agreement with the calculated results.

It has been observed that the incident angle, intensity and pulse width of the incidence pulse influenced on the spectrum and energy of CCE. To find out the influence of the BBO crystal thickness on CCE, a thinner 2.5 mm-thick BBO was put to replace the 5 mm-thick one. The blue-green CE also arose in normal incidence but was not accompanied by the yellow CE. Compared with the emission produced by the thicker BBO crystal, the blue-green CE had an obvious lower intensity and a smaller angular width, while its conical angle maintained unchanged. As MI gain is proportional to the crystal thickness, thinner crystal supplies lower efficiency of parametric wave mixing. Therefore the yellow CE hardly emerges and blue-green CE shows a lower spectral intensity with a smaller angular width. In our recent experiment ref. [17] of CCE induced by harmonic second pulses, the peak wavelength of green CE with a thinner BBO crystal revealed a red shift of 50 nm compared with the thicker one. However, in this experiment the spectral change of the blue-green CE revealed only several nanometers red shift compared with the thicker crystal, which can be seen in Fig. 8. We believe that the thickness of BBO crystal will affect the frequency shift $\delta \omega$ directly, while the influence of the crystal thickness on frequency shift diminishes as the photon pairs at $2 \omega \pm \delta \omega$ are born by the process of sum-frequency of pump fundamental photons at $\omega$ with off-axis photon pairs at $\omega \pm \delta \omega$.

3. Conclusion

In summary, bright blue-green CE has been observed experimentally in BBO crystals with different thicknesses pumped by intense femtosecond pulses at 800 nm, and the yellow CE has been observed around the blue-green CE for the first time. The influence of sum-frequency on the conical angle has been taken into account compared with ref. [16], and the calculated beam angular spectra were agreement with the experimental observations. The BBO crystal thickness seems not to affect the blue-green CE spectrum so much in experiment, while we still believe that this influence would be more pronounced in a much thicker crystal.

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References