Multicolored sideband generation based on cascaded four-wave mixing with the assistance of spectral broadening in multiple thin plates

Peng Wang,¹ Jun Liu,^{1,2,*} Fangjia Li,¹ Xiong Shen,¹ and Ruxin Li^{1,2}

¹State Key Laboratory of High Field Laser Physics, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800, China

²IFSA Collaborative Innovation Center, Shanghai Jiao Tong University, Shanghai 200240, China *Corresponding author: jliu@siom.ac.cn

Received June 16, 2015; revised July 14, 2015; accepted July 14, 2015; posted July 17, 2015 (Doc. ID 243244); published August 13, 2015

The generation of multicolored sidebands with the spectrum from 377 to 970 nm in a 0.5-mm-thick N-WG280 Schott glass based on a cascaded four-wave mixing (CFWM) process is demonstrated. The experimental setup is compact and economical. A pulse with a broadened spectrum from 670 to 900 nm is generated by utilizing two 0.18-mm-thick fused silica glass plates and is used to provide two input beams for the CFWM process. The new frequency components generated from the self-phase modulation effect in the two thin glass plates contribute to the broadening of the total spectral range of the generated multicolored sidebands. © 2015 Chinese Laser Press

OCIS codes: (190.4380) Nonlinear optics, four-wave mixing; (190.7110) Ultrafast nonlinear optics. http://dx.doi.org/10.1364/PRJ.3.000210

1. INTRODUCTION

The generation of multicolored femtosecond sidebands with central wavelengths from the UV to near-IR in bulk solid-state media by using cascaded four-wave mixing (CFWM) has been well explored since the 2000s [1-16]. In comparison with the well-developed noncollinear optical parametric amplifier (NOPA), the CFWM process can generate multicolored femtosecond sidebands at different wavelengths simultaneously [17]. In the CFWM process, when two input beams at different central wavelengths are focused on a thin transparent bulk medium with a small crossing angle, multicolored sidebands will be appeared on both sides of the incident beams. The central wavelengths of the generated multicolored sidebands can be tuned by changing the crossing angle or delay time of the two input beams. These wavelength-tunable multicolored ultrashort laser pulses have potential applications in femtosecond spectroscopy and microscopy [18,19]. To achieve as many multicolored sidebands and as broad spectra of the sidebands as possible, it is necessary to have two broadband input pulses with appropriate central wavelengths. The generation of multicolored sidebands directly after a broadband Ti:sapphire amplifier seems to be simple [1,2]. However, in most cases, the spectrum of the output pulse from a laser system directly is not broad enough. In this case, NOPA [3,4], a hollow fiber compression system [5–8], supercontinuum generation in a thick sapphire plate [9–11], self-phase modulation (SPM) in a thick glass [12], or filamentation in air [13] is adopted to extend and broaden the spectrum of the incident pulse for the CFWM process. Among them, SPM in a thick glass [12] seems to be the simplest method. However, the spectrum is not well broadened in that case due to the self-focusing effect in thick glass. Recently, separated multiple thin plates were used to broaden the spectrum of intense femtosecond laser pulses, and the output after the plates had a broadband spectrum and a nice spatial mode [20]. This kind of spectral broadening method would be attractive in the CFWM process with a narrowband input pulse. On the other hand, many bulk media have been used for multicolored sideband generation, such as tellurite glass [1], sapphire plate [2,5,13], BK7 glass [3,11], PbWO₄ [4], beta-barium borate (BBO) [6,9,10], fused silica glass [7], and diamond plate [8,12]. A transparent bulk medium with higher third-order nonlinearity can extend the CFWM process to the condition with a limitation of input intensity.

In this paper, two separated 0.18-mm-thick fused silica plates are used to broaden the spectrum of a 35 fs incident pulse at 806 nm (760–840 nm) from a Ti:sapphire amplifier. A broadband spectrum from 670 to 900 nm is generated with this compact and economical setup. Moreover, a 0.5-mm-thick N-WG280 Schott glass, which has higher third-order nonlinear susceptibility in comparison with normal glasses, is used as the nonlinear medium for generation of multicolored sidebands. As a result, the generated sidebands have a broad spectrum range that extends from 377 to 970 nm. The result proves that the use of separated multiple thin plates to broaden the spectrum of incident pulses is an effective way in the CFWM process to generate multicolored sidebands for a narrow bandwidth incident pulse.

2. EXPERIMENT

The experimental setup for multicolored sideband generation is shown in the Fig. 1. A Ti:sapphire femtosecond laser (Legend Elite, Coherent), which delivers 35 fs, 806 nm laser pulses at a repetition rate of 1 kHz, is used in the experiment.



Fig. 1. Schematic of the experimental setup. CM1, CM2, chirped mirrors; VNDF, variable neutral density filter; DM, dichroic mirror; GP, 0.5-mm-thick N-WG280 Schott glass plate.

The input beam is focused and then collimated by two spherical lenses (SLs) with focal lengths of 700 and 300 mm. Two 0.18-mm-thick fused silica plates are placed around the waist of the focused beam to generate intense spectrum-broadened pulses. The central zone of the generated ring-structured beam after the two thin fused silica plates is isolated by an iris aperture. Two chirped mirrors are used to compensate for the dispersion of the pulse. There are three bounces on each chirped mirror and each bounce would provide a dispersive compensation of -40 fs^2 . After being focused by a third SL with focal length of 1000 mm, the beam is split by a dichroic mirror with cutoff wavelength of 800 nm. The reflected part (hereafter referred to as beam1) with wavelength above 800 nm is sent to a delay line and finally overlapped with the transmitted part (hereafter referred to as beam2) with wavelength shorter than 800 nm on a 0.5-mm-thick N-WG280 Schott glass plate placed around the focus of the third SL. The energy per pulse of beam1 and beam2 after passing through the N-WG280 Schott glass plate are 7.1 and 22.6 µJ, respectively, monitored by using a silicon sensor powermeter (PM100D, Thorlabs), and the beam diameters on the N-WG280 Schott glass are 200 and 250 µm, respectively, measured by a CCD camera (BC106-VIS, Thorlabs).

3. RESULTS AND DISCUSSION

The energy of the pulse incident on the first 0.18-mm-thick fused silica plate is 121 µJ and decreases to 43 µJ after the iris aperture. By carefully controlling the position of the two thin plates around the beam waist, the beam exiting from the second thin fused silica plate acquires a bright round central zone surrounded by multiple red rings, as shown in the inset of Fig. 2. The fused silica plates are so thin that the pulse can exit the plates before the occurrence of adverse effects such as multiple filamentation and optical damage [20]. The generated broadband laser spectrum extends from 670 to 900 nm, as measured by a spectrometer (USB4000, Ocean Optics) with integration time of 100 ms after the iris aperture. From the comparison of the spectra before and after the thin fused silica plates in Fig. 2, it is obvious that the broadening of the spectrum is effective and the central wavelength is a little blueshifted. In addition, Fig. 3(a) exhibits the measured spectral stability of the broadened spectrum in half an hour. The stabilities of the spectral intensity at the 750, 800, and 850 nm are shown in Fig. 3(b) and their standard deviations are 5.9% RMS, 1.5% RMS, and 5.5% RMS, respectively. It needs to be noted that the output laser power stability from our Ti:sapphire system is about 0.75% RMS right now. A more



Fig. 2. Spectra of the pulse before and after the two thin fused silica glass plates. The inset is a photograph showing the ring-structured beam projected on a white paper behind the second thin fused silica glass plate.

stable broadened spectrum is expected to be obtained with better incident laser parameters [20]. The intense pulses with broadband spectra are good sources for multicolored sideband generation based on CFWM.

Colorful sidebands emerge when the input pulses are overlapped spatially and temporally by adjusting two reflective mirrors before the N-WG280 Schott glass and varying the delay line. Since the spectra of beam1 and beam2 are broad enough, frequency components originating from SPM in two thin fused silica plates can be chosen to participate in the CFWM process by carefully adjusting the crossing angle. When beam1 and beam2 have a crossing angle of 3° in air, the multicolored CFWM sidebands are generated and projected on a white sheet of paper and a near-IR card that are placed 300 mm after the N-WG280 Schott glass plate, as Fig. 4(a) shows. More than nine frequency upconversion (FUC) sidebands on a white paper and one frequency downconversion (FDC) sideband on a NIR card can be observed. The colors of the sidebands change gradually from red to deep blue. The higher order FUC sidebands are spatially well separated, but relatively smaller and weaker. When we remove the two 0.18-mm-thick fused silica plates for spectral broadening, only a few weak sidebands are generated. Even though



Fig. 3. (a) Spectral stability of the broadened spectrum in half an hour. (b) The spectral intensity stabilities at 750, 800, and 850 nm in half an hour.



Fig. 4. (a) Photo of the multicolored sidebands when two thin fused silica plates are used to broaden the spectrum and an N-WG280 Schott glass is used as the nonlinear medium. (b) Photo of the sidebands when the two 0.18-mm-thin fused silica plates for spectral broadening are removed and the input power is increased. A NIR card is placed on the left of the input beams.

the input power is increased to generate more sidebands, as shown in the Fig. 4(b), the color change of the sidebands is barely recognized, which indicates a narrow total spectral range of the sidebands. As a result, the spectrum broadening in two thin fused silica glass plates is important in multicolored sideband generation for a narrow bandwidth incident pulse. We also use a 0.5-mm-thick CaF_2 plate, which has a relatively lower third-order nonlinear susceptibility, to substitute the N-WG280 Schott glass and maintain the same input power. The color of the sidebands is weakened, and no more than three sidebands are generated. This result indicates the use of an N-WG280 Schott glass with a higher third-order nonlinear susceptibility makes it possible to generate multicolored femtosecond pulses with low input intensity, which would be useful when the incident pulse energy is limited.

The spectra of the input beams and the separated CFWM sidebands generated in the N-WG280 Schott glass plate with crossing angle of 3° in air and shown in Fig. 4(a) are picked up by using a spectrometer (USB4000, Ocean Optics), as shown in Fig. 5. The spectrum range of the sidebands extends from 377 to 970 nm, which is comparable with the multicolored sideband generation when a hollow fiber compression system is used [5–8]. Each spectrum of the shown sidebands has a nice Gaussian-like spectral profile, even though the spectra of the input beams are modulated because of the SPM process. The full width at half-maximum (FWHM) spectral bandwidths of the sidebands decrease from 47 to 8 nm with the increase of the order number of the sidebands. The



Fig. 5. Spectra of input beams and the generated sidebands. The nine spectra on the left side belong to the FUC sidebands and the spectrum on the right side belongs to the FDC sideband. The spectra of the input beams are represented by the dotted lines.



Fig. 6. Central wavelengths of the sidebands change with the crossing angle.

carrier-envelope phase (CEP) of each sideband is stable if only the original driving pulse from the amplifier is CEP stabilized.

The output powers of the generated sidebands are monitored by using a silicon sensor powermeter (PM100D, Thorlabs). The energy of the first-order FUC signal reaches 0.121 μ J and its standard deviation is 2.6% RMS in 7 min. By controlling the fluctuation of temperature and air flowing in our laboratory carefully, the stabilities of the sidebands should be improved. By measuring the powers of beam1 and beam2 before and after multicolored sideband generation after the N-WG280 Schott glass plate, the energy conversion to the sidebands is calculated to be about 1%. The limited energy of the new frequency components originating from the SPM process lead to the relatively lower energy conversion in the present proof-of-principle experiment.

In the CFWM process, the phase-match condition should be satisfied: $(m + 1) \omega_2 - m\omega_1 = \omega_{FUm}$ and $k_{FUm} - (k_2 - k_1) = k_{FU(m-1)}$. In the equations, k_1 and k_2 are wave vectors of beam1 and beam2 with respective angular frequencies of ω_1 and ω_2 , while k_{FUm} and $k_{FU(m-1)}$ are wave vectors of the generated *m*th-order and (m - 1)th-order FUC sidebands. The central wavelengths of the FUC sidebands from the first order to the ninth order are calculated to be 647, 591, 544, 504, 469, 439, 413, 389, and 368 nm, which are in good accordance with the experimental results.

Since the input beams have broadband spectra, the frequency components involved in the CFWM process can be chosen by changing the crossing angle. When we increase the crossing angle to 4.5° in air, fewer sidebands are generated. However, the central wavelengths of the sidebands are obviously changed, as shown in the Fig. 6.

4. CONCLUSION

In summary, we have demonstrated the generation of multicolored sidebands with spectra from 377 to 970 nm in a 0.5-mm-thick N-WG280 Schott glass based on the CFWM process. The SPM process in two 0.18-mm-thick glass plates is used to broaden the spectrum of an intense laser pulse and make the experimental setup compact and economical. This method can be extended to other wavelengths with narrow bandwidths. The generated sidebands have potential applications in many fields, such as femtosecond spectroscopy and microscopy [11].

ACKNOWLEDGMENT

This work is supported by the National Natural Science Foundation of China (NSFC) (grants 61178006, 11274327 and 61221064), and the Recruitment Program of Global Youth Experts.

REFERENCES

- H. Zhang, Z. Zhou, A. Lin, J. Cheng, H. Liu, J. Si, F. Chen, and X. Hou, "Controllable cascaded four-wave mixing by two chirped femtosecond laser pulses," Appl. Phys. B 108, 487–491 (2012).
- P. Wang, J. Liu, F. J. Li, X. Shen, and R. X. Li, "Generation of highenergy tunable multicolored femtosecond sidebands directly after a Ti:sapphire femtosecond laser," Appl. Phys. Lett. 105, 201901 (2014).
- H. Crespo, J. T. Mendonca, and A. Dos Santos, "Cascaded highly nondegenerate four-wave-mixing phenomenon in transparent isotropic condensed media," Opt. Lett. 25, 829–831 (2000).
- K. Wang, M. C. Zhi, X. Hua, J. Strohaber, and A. V. Sokolov, "Coherent broadband light generation with a double-path configuration," Appl. Opt. 53, 2866–2869 (2014).
- J. Liu and T. Kobayashi, "Cascaded four-wave mixing and multicolored arrays generation in a sapphire plate by using two crossing beams of femtosecond laser," Opt. Express 16, 22119–22125 (2008).
- J. Liu, J. Zhang, and T. Kobayashi, "Broadband coherent anti-Stokes Raman scattering light generation in BBO crystal by using two crossing femtosecond laser pulses," Opt. Lett. 33, 1494–1496 (2008).
- J. Liu and T. Kobayashi, "Wavelength-tunable, multicolored femtosecond-laser pulse generation in fused-silica glass," Opt. Lett. 34, 1066–1068 (2009).
- C.-H. Lu, L.-F. Yang, M. Zhi, A. V. Sokolov, S.-D. Yang, C.-C. Hsu, and A. H. Kung, "Generation of octave-spanning supercontinuum by Raman-assisted four-wave mixing in single-crystal diamond," Opt. Express 22, 4075–4082 (2014).
- 9. W. Liu, L. Zhu, and C. Fang, "Observation of sum-frequencygeneration-induced cascaded four-wave mixing using two crossing femtosecond laser pulses in a 0.1 mm beta-bariumborate crystal," Opt. Lett. **37**, 3783–3785 (2012).

- W. Liu, L. Zhu, L. Wang, and C. Fang, "Cascaded four-wave mixing for broadband tunable laser sideband generation," Opt. Lett. 38, 1772–1774 (2013).
- L. Zhu, W. Liu, and C. Fang, "Tunable sideband laser from cascaded four-wave mixing in thin glass for ultra-broadband femtosecond stimulated Raman spectroscopy," Appl. Phys. Lett. 103, 061110 (2013).
- J. He, J. Du, and T. Kobayashi, "Low-threshold and compact multicolored femtosecond laser generated by using cascaded four-wave mixing in a diamond plate," Opt. Commun. 290, 132–135 (2013).
- P. Wang, J. Liu, F. J. Li, X. Shen, and R. X. Li, "Filamentation assisted generation of tunable multicolored femtosecond sidebands based on cascaded four-wave mixing," Laser Phys. 25, 055401 (2015).
- J. Liu and T. Kobayashi, "Generation and amplification of tunable multicolored femtosecond laser pulses by using cascaded four-wave mixing in transparent bulk media," Sensors 10, 4296–4341 (2010).
- M. Zhi, X. Wang, and A. V. Sokolov, "Broadband light generation in Raman-active crystals driven by femtosecond laser fields," in *Advances in Lasers and Electro Optics* (InTech, 2010), pp. 655–682.
- T. Kobayashi, J. Liu, and Y. Kida, "Generation and optimization of femtosecond pulses by four-wave mixing process," IEEE J. Sel. Top. Quantum Electron 18, 54–65 (2012).
- G. Cerullo and S. De Silvestri, "Ultrafast optical parametric amplifiers," Rev. Sci. Instrum. 74, 1–18 (2003).
- M. Levenson, Introduction to Nonlinear Laser Spectroscopy (Elsevier, 2012).
- P. Török and F. J. Kao, Optical Imaging and Microscopy (Springer, 2007).
- C.-H. Lu, Y.-J. Tsou, H.-Y. Chen, B.-H. Chen, Y.-C. Cheng, S.-D. Yang, M.-C. Chen, C.-C. Hsu, and A. H. Kung, "Generation of intense supercontinuum in condensed media," Optica 1, 400–406 (2014).