Cascaded four-wave mixing and multicolored arrays generation in a sapphire plate by using two crossing beams of femtosecond laser

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Abstract: Ultrabroadband signals were found to be generated by cascaded four-wave mixing in a bulk medium by using two crossing femtosecond laser pulses. As many as fifteen spectral upshifted pulses and two spectral downshifted pulses were obtained with spectral bandwidth broader than 1.5 octaves. Bright upshifted pulses were observed on both sides of the two crossing beams. Two dimensional multicolored arrays were observed for the first time in a sapphire plate medium.

OCIS codes: (190.4380) Nonlinear optics, four-wave-mixing; (320, 2250) Femtosecond phenomena; (190, 7110) Ultrafast nonlinear optics.

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#100646 - $15.00 USD Received 26 Aug 2008; revised 12 Oct 2008; accepted 25 Oct 2008; published 19 Dec 2008

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1. Introduction

Nowadays, optical parametric process based on three-wave mixing has been well established and using this process in second-order nonlinear crystal tunable ultrashort pulse has been widely generated across wide spectral range [1]. By using a noncollinear phase matching method, sub-two-cycle ultrashort pulse was generated in visible [2] and NIR [3] region through optical parametric amplification in a BBO crystal.

Recently, ultrabroadband spectrum and ultrashort pulse generated through four-wave-mixing (FWM) driven by the third-order susceptibility became a hot research spot. Tunable ultrashort pulses in visible was generated in a gas cell by FWM in filamentation [4]. Fuji and Suzuki also reported that few cycle ultrashort pulses in deep ultraviolet and mid-infrared also were generated by FWM through filamentation in gases last year [5, 6]. In bulk solid-state media, due to high material dispersion, phase matching will be obtained only if the pump beams have a finite crossing angle. When the two crossing beams were femtosecond laser pulses, cascaded FWM sidebands signals occurred in a piece of BK7 glass [7]. In some Raman-active nonlinear crystals, different-frequency resonant FWM named coherent Stokes and anti-Stokes Raman scattering (CSS and CARS, respectively) takes place and broadband sidebands high-efficiency generation was reported [8-13]. High efficiency and high-energy noncollinear four-wave optical parametric amplification in a transparent bulk Kerr medium was also obtained by using ps pump pulse [14-15]. In comparison with a conventional phase-matching three-wave mixing process, FWM are more widely applicable to nearly all transparent media in broad difference wavelength range. It is because of FWM phase matching requirement is more flexible due to more frequencies are involved and of non-requirement of symmetry in nonlinear materials.

In this paper, broadband cascaded FWM lasing were observed, the spectra of the output signals extended from UV to near infrared with more than 1.5 octaves. Bright up-conversion signal appear on both sides of the input beams. As far as we know, bright two-dimensional multicolored arrays from a sapphire plate medium were observed for the first time.

2. Experiment setup

The experimental setup was sketched in Fig. 1. A femtosecond regenerative amplifier system (Micra+Legend-USP) was used as a pump source. The output pulse of the system is centered at 800nm, with 40fs pulse duration, 1kHz repetition rate and 2.5W average power. Laser pulse after the regenerative amplifier system was split into two beams by using a beamsplitter. One beam (beam1) was spectrum broadened in a hollow fiber with 250μm inner diameter, 60cm length that filled with krypton gas. The broadband spectrum after the hollow fiber was dispersion compensated with a chirped mirror pair and a pair of glass wedge. The pulse duration after the hollow fiber compressor was about 10fs. Another beam (beam2) passed through a delay stage with less than 3-fs resolution. Beam2 was first attenuated by a variable neutral density (VND) filter and then focused into a 2-mm thick sapphire plate with a lens. Beam1 was focused into the sapphire plate directly with a concave mirror. An optical fiber was used to pick up different order signal to measure their spectrum by using a multichannel spectrometer (USB4000 and NIR256-2.5, Ocean Optics). The optical fiber was fixed on an

#100646 - $15.00 USD Received 26 Aug 2008; revised 12 Oct 2008; accepted 25 Oct 2008; published 19 Dec 2008

arm installed on a stage, which can be moved in the direction normal to the beam. In this case, the position of the generated sidebands was recorded.

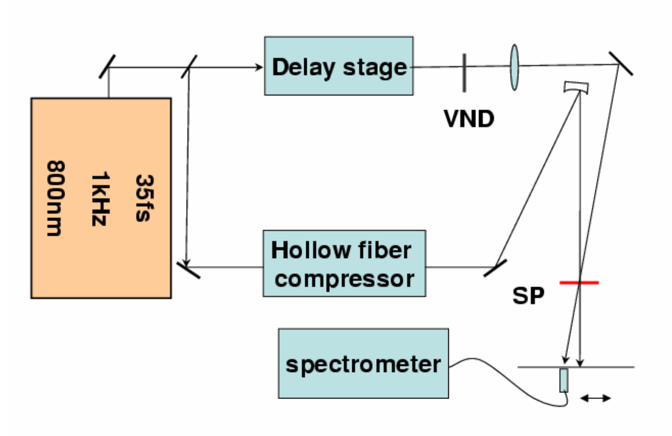

Fig. 1. Experimental setup. VND: variable neutral-density filter. SP: sapphire plate

3. Experimental results and discussion

In the experiment, the beam diameter of beam1 and beam2 on the sapphire plate were about 200µm and 800µm, respectively. The crossing angle between beam1 and beam2 on the sapphire plate was 2.0 degree. At first, a piece of 3mm-thick R850 glass filter was placed at the beam1 path to cut off the high frequency components of the pulse. The laser spectrum of beam1 after the R850 glass filter is shown in Fig. 2 as blue solid line. The spectrum extends from 800nm to 920nm. The magenta color solid line also shows the spectrum of beam2 with about 30nm FWHM spectral bandwidth. The pulse energy after the R850 glass was 9µJ and the pulse energy of beam2 was 195µJ. Bright sidebands only appeared at the side of beam2 when beam1 and beam2 overlapped well both in space and time. Photograph of sidebands light was taken on a white sheet of paper which was placed behind the sapphire plate.

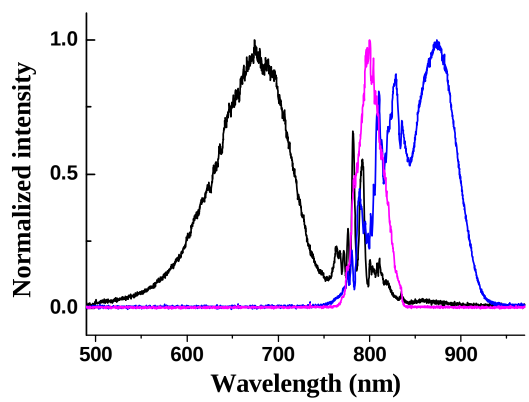

Fig. 2. The spectra of beam2 (magenta line) and beam1 with a R850 band pass glass filter around 850nm(blue line) or a cut off glass filter at 820nm (black line).

Figure 3(a) shows the photograph of the sideband signals. As many as 15 upshifted sidebands and two downshifted sidebands were generated. The spectra of the sidebands extended from UV to near infrared with more than 1.5 octaves. Figures 3(b) and (c) show the

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spectra of the sidebands from UV to the near infrared. To make it clearer, the spectra were shown in two figures. The second-order downshifted spectrum (R2) in near infrared was measured by an infrared spectrometer (NIR256-2.5, Ocean Optics). The spectral bandwidths of the low order uphshifted and downshifted sidebands were even broader than that of the pump beam (beam2). It means that the spectra of the sidebands are dependent on the spectral bandwidth of the two input pulses. The spectrum of the low order sidebands also exhibits an obvious interference pattern. An enlarged top part of AR1 spectrum showed in Fig. 3(d). If the spectra of the two input pulses were broad enough, different pairs with different frequency components from each of the input pulses can generate the same spectrum in the sidebands. Then the interference pattern will appear when the delay between the pair component is short enough. The spectral shapes of the high-order upshifted sidebands are very similar to each other, as shown in Fig. 3(b). The spectral FWHM bandwidths of the high order upshifted sidebands are about 410cm^{-1} and more or less similar to each other. The frequency space between two neighbor sideband was decreased from 1203cm^{-1} (AR1 and AR2) to 625cm^{-1} (AR14 and AR13) as the order increased. In the experiment, the brightness and number of the sidebands were reduced when we decrease the pulse energies of beam1 or beam2.

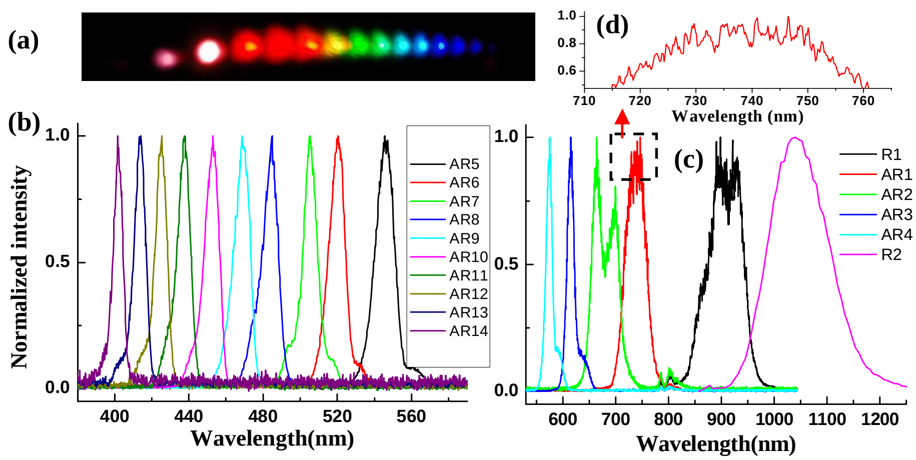

Fig. 3. (a) Photograph and (b)spectra of the high-order sidebands and (c) the low-order sidebands (d) enlarged peak part of spectrum of AR1 observed when a 3mm-thick R850 glass filter was used in beam1 path. AR*m* (*m*=1-14) refers to the *m*th-order upshifted spectrum, and R*n* (*n*=1, 2) refers to the *n*th-order downshifted spectrum.

In order to study the short wavelength range of the laser spectrum we replaced the R850 glass filter with another short wavelength pass filter, which cut at 820nm. The pulse spectrum after the filter is also shown in Fig. 2 with a black line. The spectrum extends from 600nm to 820nm. The pulse energy after the filter glass was about 40μJ. The pulse energy of beam2 was about 115μJ. In this case, bright sideband lights appeared only on the side of beam1 even the intensity of beam2 on the sapphire plate was higher than that of beam1. Photographs of sidebands light on a white sheet of paper placed behind the sapphire plate were shown at the top of Fig. 4(a). Together with the two experiments, it was found that bright sidebands always appeared on the side of the beam with high frequency. More upshifted sidebands than downshifted sidebands appear. This asymmetry between the upconversion and downconversion processes due to phase matching [7]. There was a bright beam A1 between beam1 and beam2, which was a little strange at first sight. As we can see, the sideband overlapped slightly with beam2 in the spectrum and in space, as shown in Fig. 4(a). This phenomenon is explained in term of a cascade FWM signal between beam1 and AR1. In the process, the first order FWM signal AR1 was generated between beam1 and beam2, and then

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cascaded FWM between beam1 and AR1 will be generated upshifted beam AR2 and downshifted beam A1. Both the spectrum and space shift due to the phase matching condition.

The number of sidebands was not as many as that on the side of beam2. However, the spectrum of the sidebands almost extends to the same spectral range resulting in more than 1 octave. The spectral bandwidth of the sideband was not as broad as the seed from beam1. This is because that the spectrum of the sidebands dependent not only on the spectrum of the pump beam but also on the phase matching condition. The spectrum and direction from the sapphire plate of the sideband were dependent on the crossing angle between beam1 and beam2. When the crossing angle was smaller, the gap of space and spectrum between neighboring sidebands were narrower. The intensity of the sidebands has optimum crossing angle. In the experiment, when the crossing angle was about 2 degrees, the sidebands were brightest. The conversion efficiency from the pump energy to the sum of the sideband energies was about 12%. We also changed the polarization of beam2 normal to beam1, very weak few sidebands were observed. These properties were nearly the same as those reported previously [9, 10].

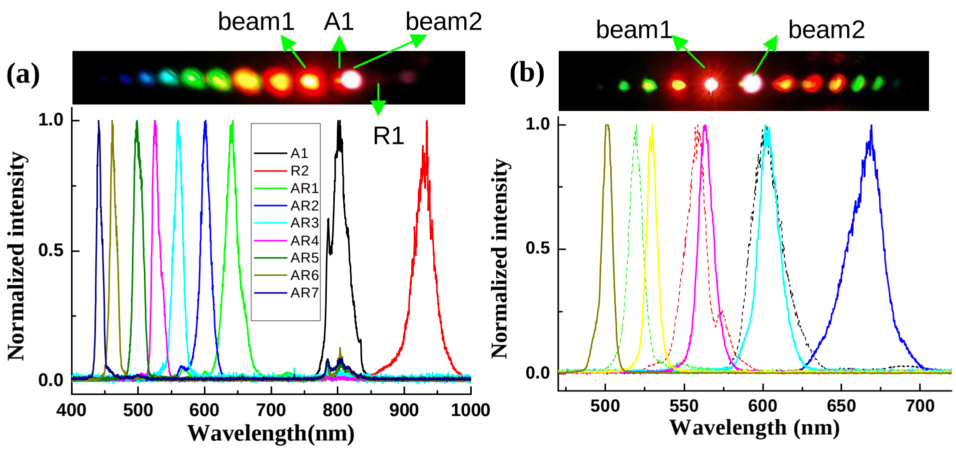

Fig. 4. Photographs and spectra of the sidebands light (a) when a short-wavelength-pass glass filter with a cut wavelength at 820nm was used in beam1 path; (b) when no glass filter was used in beam1 path. AR*m* (*m*=1-7) refers to the *m*th-order upshifted spectrum, and R*n* (*n*= 2) refers to the *n*th-order downshifted spectrum.

In the experiment, when there was no filter inserted in the beam1 path, the spectrum of beam1 extends from 600nm to 920nm. Super-continuum light will be generated in this case due to high intensity on the sapphire plate. Therefore, we replaced the concave mirror with another concave mirror with different focal length to let the beam diameter of beam1 on the sapphire plate is about 800μm which fit to the diameter of beam2. The pulse energy of beam1 was about 55μJ. When the input pulse energy of beam2 is increased to about 195μJ, bright upshifted sidebands appeared on the both sides of beam1 and beam2 at the same time. In the experiment, to make sidebands to be separated clearly, the crossing angle between beam1 and beam2 on the sapphire plate was set at 2.75 degree. The photograph of the sidebands light on a white sheet of paper placed behind the sapphire plate was shown at the top of Fig. 4(b). The spectra of the sidebands were shown as in Fig. 4(b). The dotted lines and solid lines show spectra of sidebands on the sides of beam1 and beam2, respectively. The spectral wavelength of the sidebands of the same order on both sides was different from each other. Even though the spectra of the first-order signal with black dotted line and the second-order signal with cyan solid line are nearly overlapping, the spectra of high order were shifted to each other as the order increases. The spectral bandwidth of the sidebands with dotted line was a little broader than that of the near sidebands with solid line. Since the spectrum of the sideband was

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dependent on the crossing angle, there is a possibility to obtain much more separated spectrum sidebands by optimizing the crossing angle between beam1 and beam2.

In the experiment, the same phenomenon was observed when a piece of 1-mm thick fused silica glass was used. Very recently, near-single-cycle 2.7fs visible-UV pulse was reported by Fourier synthesis broadband cascaded four-wave mixing signals generated from bulk silica [16]. These kinds of sidebands can be used as the light source for multicolored pump-probe experiment and as the seed for multicolor laser amplification. Sub-femtosecond pulse may be able to be obtained by this method.

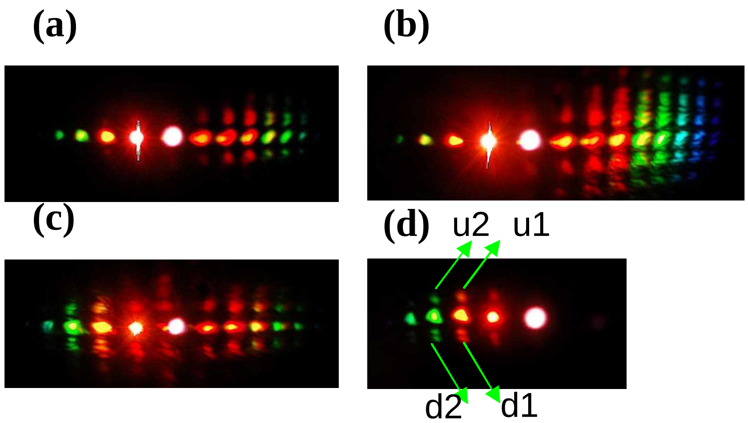

Fig. 5. Photographs of the multicolored arrays under several different conditions. The conditions were, (a) pulse energy of beam2 was 220µJ; (b) pulse energy of beam2 was increased to 250µJ; (c) the delay of beam2 was tuned about 7fs at 250µJ input pulse energy; and (d) a short wavelength pass glass filter cut at 820nm was inserted in the beam1 path.

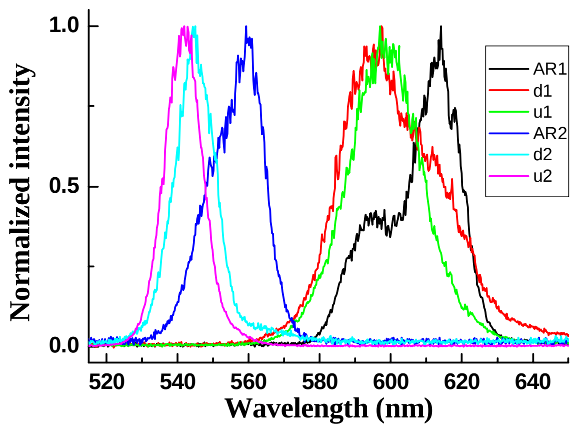

Fig. 6. The spectra of different laser spots shown in Fig. 5(d). AR1: the spot beside beam1 on same line, d1: the spot below AR1, u1: the spot over AR1. AR2: the second order upshifted beam, d2: the spot below AR2, u2: the spot over AR2.

Then, we increased the pulse energy of the beam2 by rotating the VND filter in beam2 path. As the pulse energy was increased to 220µJ, two-dimensional (2D) multicolored arrays were observed clearly in the experiment, as shown in Fig. 5(a). The 2D multicolored arrays only appeared on the right of input beams. These 2D arrays signals show periodic two-dimensional lattice structure. When the pulse energy of beam2 was increased to 250µJ, much

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more lines and brighter multicolored arrays were observed, as shown in Fig. 5(b). The multicolored arrays only existed on the side of beam2. It maybe due to two input beams did not absolutely normal to the sapphire plate that the multicolored arrays were slightly unsymmetrical. As we tuned the delay of beam2 about 7fs, multicolored arrays appeared on the both sides of beam1 and beam2. Figure 5(c) shows the photograph of the multicolored arrays on the both sides of beam1 and beam2. This is because that there were a lens and a VND filter in the beam2, which will induce positive chirp to the beam2. Then, a short wavelength pass glass filter cut at 820nm was inserted in the path of beam1, multicolored arrays again were observed, as shown in Fig. 5(d). By varying the intensity, delay or polarization of one input beam, the two-dimensional multicolored array can be controlled. It needs to note that the sapphire plate was not damaged in the whole experiment.

In the experiment, we measured the spectrum of the multicolored array on different line and the same row. The u1 and u2 refer spots over AR1 and AR2, The d1 and d2 refer spots below AR1 and AR2, as shown in Fig. 5(d). Figure 6 shows the spectra of laser spots at the row of AR1 and AR2 besides beam1. The spectra of u1, u2, d1 and d2 were shifted to the shorter wavelength by about 20nm with respect to that of the spot on the center line. This spectral shift is considered to be due to different angle between them to satisfy the phase matching condition. The 2D multicolored arrays may be used for all-optical switch of two-dimensional laser beam array.

Since the multicolored array pattern was very sensitive to the delay between two input beams, transient cascade FWM is considered to play a domain effect rather than Raman scattering which was less sensitive to the delay time because of the vibrational dephasing time being in the order of picoseconds. Some researches showed that FWM processes play an important role in multicolor solitons generation [17, 18]. Similar two-dimensional multicolored arrays were observed in a quadratic nonlinear crystal of BBO by H. Zeng et al. [19] owing to cascaded noncollinear quadratic processes. Very recently, two-dimension multicolored arrays due to the Raman scattering and FWM both due to the third-order nonlinearity and six-wave-mixing due to the fifth order nonlinear processes were also observed when three beams were focused into a diamond [10]. The 2D multicolored array may be used for all-optical switching device of two-dimensional laser beam array.

4. Conclusion

In conclusion, we experimentally report ultrabroadband signals generated in a bulk medium by using two crossing femtosecond laser pulses. Spectra of the sidebands extend from UV to NIR with bandwidth broader than 1.5 octaves. Bright upshifted pulses were observed on the both sides of the two crossing beams at the same time when the input pulses owe broadband spectrum. Two-dimensional multicolored arrays were observed for the first time in a piece of sapphire plate.

Acknowledgment

We thank Zhiguang Wang for his technical assistance. This work was partly supported by the 21st Century COE program on “Coherent Optical Science” and partly supported by the grant from the Ministry of Education (MOE) in Taiwan under the ATU Program at National Chiao Tung University. A part of this work was performed under the joint research project of the Laser Engineering, Osaka University, under contract subject B1-27.

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