Low threshold power density for the generation of frequency up-converted pulses in bismuth glass by two crossing chirped femtosecond pulses

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Abstract: We investigated the generation of frequency up-converted femtosecond laser pulses by nondegenerate cascaded four-wave mixing (CFWM) in a bismuth-oxide glass (BI glass). Broad-bandwidth light pulses with different propagation directions were simultaneously obtained by using two small-angle crossing femtosecond laser pulses in BI glass. Experimental results show that the threshold power density for the generation of broad-bandwidth femtosecond pulses in BI glass is one order of magnitude lower than that in fused silica.

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1. Introduction

The cascaded four-wave mixing (CFWM) in femtosecond time scale was firstly reported in BK7 glass [1], in which the multicolor frequency up-converted femtosecond laser pulses were observed. In the case of two ultrashort laser pulses propagating through transparent isotropic condensed media, phase mismatch is partly compensated by interacting at a finite crossing angle [2]. Because of the broad spectral range involved, the phase matching condition in CFWM process is very flexible.

Recently, broadband CFWM signals driven by the third-order susceptibility have been experimentally generated in sapphire plate [3], and fused silica [4,5]. These generated tunable multicolor ultrashort pulses can be used in many fields, such as multicolor pump-probe experiments [6–8], high-power single cycle pulse synthesis [9,10], femtosecond coherent anti-Stokes Raman spectroscopy (CARS) [11,12], and measurements with high time resolution [13–15]. However, high energy incident pulses should be used due to small nonlinear refractive-index n_2 of the above-mentioned isotropic media. It limits the application of CFWM technique in the future.

In this paper, we investigated the generation of frequency up-converted femtosecond laser pulses by nondegenerate CFWM in a bismuth-oxide glass (BI glass). Broad-bandwidth light pulses with different propagation directions were simultaneously obtained by using two small-angle crossing femtosecond laser pulses in BI glass, demonstrating that the threshold power density is one order of magnitude lower than that in fused silica.

2. Experiments

The non-resonant-type homogeneous BI glass was prepared by melting Bi_2O_3, SiO_2, B_2O_3 according to a certain proportion of 60%, 20%, 20%, respectively. A small component of 0.15%-CeO_2 was used to suppress the precipitation of Bi metal during melting. The raw materials were mixed in a Pt crucible in a SiC furnace and melted at 1150 °C for 1.5 h in air. The melted mixtures were poured onto a stainless-steel plate at room temperature to remove strain in the glass. A linear absorption spectrum of the sample showed that the absorption edge located at 450 nm and there was no evident absorption in near-infrared region [16].

The experimental setup is shown in Fig. 1(a). The multi-pass amplified Ti:sapphire laser emitted 30 fs, 800 nm laser pulses at a repetition rate of 1 kHz. The emitted beam was split into two parts (beam 1 and beam 2) which were focused into the sample by a lens with the focal length of 300 mm at a small angle of 2.5°. The spectra of the two incident pulses are both extended from 750 to 860 nm, which are shown in Fig. 1(b). Two variable neutral density (VND) attenuators were used to change the intensities of beam 1 and beam 2, respectively. A time-delay device, which was controlled by a computer, was used to adjust the timing of pulse collisions. The time delay Δt between the two pulses was calibrated by use of the autocorrelation signal from second-harmonic generation (SHG) in a 1-mm-thick BBO crystal. The full width at half maximum (FWHM) of the incident pulse width is about 240 fs at the position of the sample. Beam 1 (k_1) arrives at the sample before beam 2 (k_2), which is referred to positive time delays. The beam diameter of beam 1 and beam 2 on the sample were about 240

and 300 μm, respectively. In our experiments, no supercontinuum was detected, and the influence of supercontinuum generation on our experimental results can be excluded. The intensity and spectral profiles of the nondegenerate CFWM signals were detected by a photomultiplier tube (PMT) and an optical multi-channel analyzer (OMA), respectively.
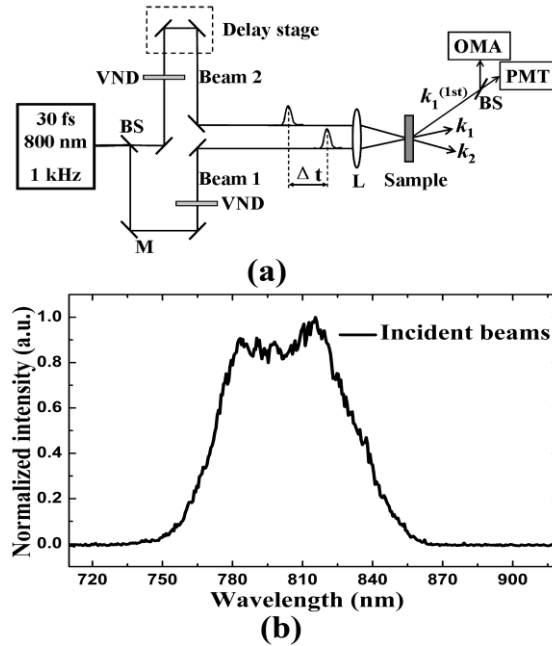

(a)

(b)

Fig. 1. (a) Experimental arrangement. BS: beam splitter; M: silver mirrors; VND: variable neutral density (VND) attenuators. (b) Normalized spectra of two incident beams.

3. Experiment results and discussion

The phase matching condition of the first order nondegenerate CFWM signal on beam 1 side can be described by $k_1^{(1st)} = 2k_1 - k_2$ at the positive time delay scale. Two k_1 photons and one k_2 photon generate the first-order frequency up-converted photon $k_1^{(1st)}$, and the generated $k_1^{(1st)}$ will take part in the next four-wave mixing process. Thereafter the higher mth-order signals on beam 1 side obey the following phase match condition [17]: $k_1^{(mth)} = k_1^{(m-1)th} + k_1 - k_2 \approx (m+1)k_1 - mk_2$. It would be expected that the nondegenerate CFWM signals would be also appeared on beam 2 side when the time delay is negative.

In the experiment, we set the intensity of the beam 1 and beam 2 at 22×10^9 and 28.5×10^9 W/cm^2, respectively. Figure 2(a) shows the photograph of the sideband signals on beam 1 side in 1 mm BI glass, in which the time delay Δt was 130 fs. Five sideband signals generated by nondegenerate CFWM were simultaneously observed. The normalized spectra of the sideband signals on beam 1 side are shown in Fig. 2(b). As many as five up-converted frequency bands (L1-L5) were obtained, whose center wavelength located at 731, 707, 680, 656, 631 nm, respectively, shifting about 25 nm for each neighboring band. The FWHM of the spectrum of each band is about 10 nm, which was much narrower than the widths of both incident beams. The wavelength blue-shift interval of neighbor orders was estimated to be about 25 nm. The transmitted differential spectra of beam 1 and beam 2 is shown in Fig. 2(c), which are the difference of the transmitted spectra of one incident beam (beam 1 or beam 2) with and without the other incident beam (beam 2 or beam 1) focusing on the same spot. The transmitted differential spectra show a valley at 761 nm for beam 1 and a valley at 809 nm for beam 2,

manifesting that a L1 photon is generated by two 761 nm photon (k_1) and one 809 nm photon (k_2).

When the incident intensities of beam 1 and beam 2 were set at 22×10^9 and 28.5×10^9 W/cm^2, the incident intensities of which were one order of magnitude smaller than that in the previous works [4], the overall energy conversion efficiency from the incident beams to the sidebands reached approximately 5%. Furthermore, the conversion efficiency may be further increased by increasing the power intensity of the two incident beams or by increasing the concentration of Bi_2O_3 in BI glass [16].
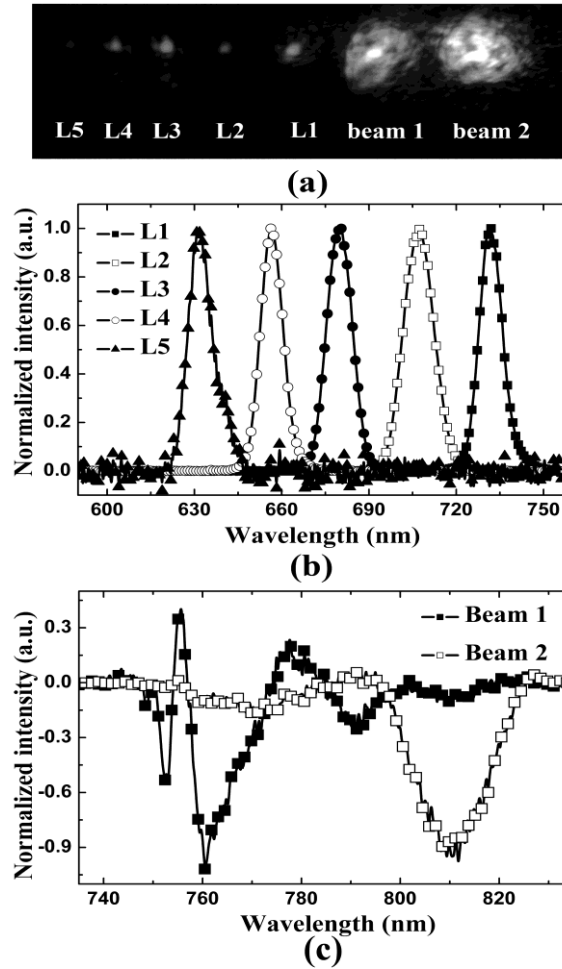

Fig. 2. (a) Photograph of the nondegenerate CFWM signals appeared on beam 1 side in 1 mm BI glass, in which the time delay Δt was 130 fs. Lm (m = 1-5) refers to the mth-order signal. (b) The normalized spectra of the sideband signals. (c) The transmitted differential spectra of beam 1 and beam 2.

Figure 3(a) shows the pattern of another five bright spots (R1-R5) which were generated simultaneously on beam 2 side at a negative time delay $\Delta t=-130$ fs. The normalized spectra of the signals are shown in Fig. 3(b), in which we can see that the center wavelength of R1, R2, R3, R4, R5 located at 734, 714, 688, 669, 645 nm, respectively. From Fig. 3(c), we can see that the primary incident pairs (k_1, k_2) in the nondegenerate CFWM processes for negative time delay were 766 and 813nm, which is different from that at positive time delay 130 fs.
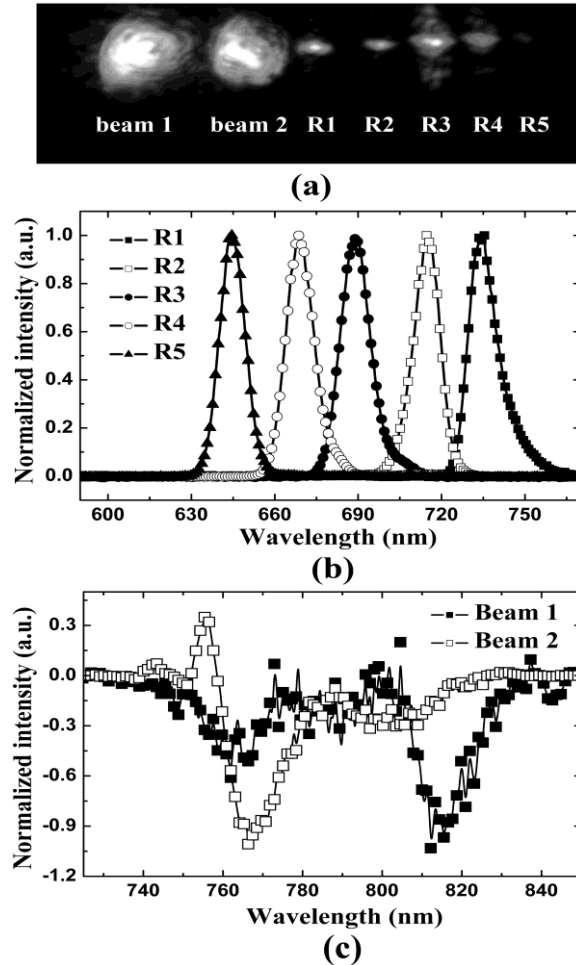
Fig. 3. (a) Photograph of the nondegenerate CFWM signals appeared on beam 2 side in 1 mm BI glass, in which the time delay Δt was −130 fs. Rm (m=1-5) refers to the mth-order signal. (b) The normalized spectra of the sideband signals. (c) The transmitted differential spectra of beam 1 and beam 2.

The center wavelength of nondegenerate CFWM signals on beam 2 side was different from that on beam 1 side for the same order. The difference of the center wavelength was 3, 7, 8, 13 and 14 nm for the first, the second, the third, the fourth and the fifth order, respectively. The difference of the center wavelength described above results from the discrepancy of the pair (k_1, k_2) participated in nondegenerate CFWM between the delay time at 130 fs and that at −130 fs. Since the incident beams are long chirped due to group velocity dispersion in lenses, the air, and the BI glass, the incident wavelengths fitting the phase matching condition for L1-L5 bands are slightly different from that for R1-R5.

It was found that the sideband signals always appeared on the side of the beam in which high frequency components took part in nondegenerate CFWM processes, no matter whether the time delay is positive or negative. Moreover, the polarization of these signals kept the same as that of the incident beams. The brightness and number of the sidebands were sensitively reduced by decreasing the input pulse energies of beam 1 and beam 2. When the two incident pulses overlapped completely on time delay scale, either of the two sides sideband signals disappeared.

In order to study the threshold energy of frequency up-converted femtosecond pulses generation, we measured the intensity dependence of L1 on beam 1 in the BI glass and a fused silica glass, respectively, where the thickness of fused silica also was 1 mm. In this measurement, we kept the intensity of beam 2 at 8.0×10^9 W/cm^2 and the time delay Δt between two incident beams at 130 fs. As shown in Fig. 4, the intensity of L1 increased monotonously with the intensity of beam 1 both in BI glass and fused silica. The threshold of beam 1 to generate the L1 in BI glass was estimated to be about 2.6×10^9 W/cm^2, while the threshold of beam 1 in fused silica was about 60×10^9 W/cm^2. The threshold of beam 1 in fused silica was one magnitude larger than that in the BI glass. The significant decreasing of threshold in BI glass was attributed to its large nonlinear refractive-index $n_2\sim2.93\times10^{-15}$ cm^2/W [18], which was one magnitude larger than that of fused silica [19].
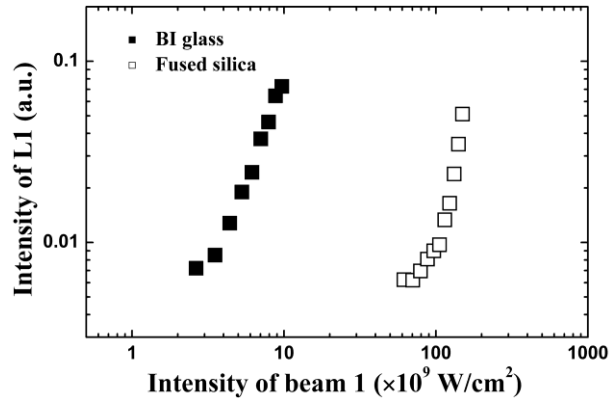

Fig. 4. Intensity dependence of L1 on the intensity of beam1 in BI glass (closed squares) and fused silica (open squares).

4. Conclution

In summary, we investigated the generation of frequency up-converted femtosecond laser pulses by nondegenerate CFWM in BI glass. BI glass exhibits low threshold in generating frequency up-converted femtosecond lasers by nondegenerate CFWM due to its large nonlinear refractive-index. Broad-bandwidth light pulses with different propagation directions can be easily generated simultaneously, which promotes the BI glass to a candidate material of ultrafast nonlinear optical devices, such as fast all-optical gate switches [20], optical wavelength converters [21].