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Nonlinear Cascaded Femtosecond Third Harmonic Generation by Multi-grating Periodically Poled MgO-doped Lithium Niobate

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ABSTRACT

Nonlinear cascaded femtosecond third harmonic generation was experimentally investigated pumped by 100 fs pulses at optical communication band 1550 nm using a multi-grating 5 mol. % MgO-doped periodically poled lithium niobate crystal. The optimized efficiency of 10.8% was achieved with the simultaneous phase-matching of the second harmonic and sum frequency process. And the third harmonic spectrum reached as broad as 8.7 nm because of the choosing of a small group velocity mismatching between the fundamental and second harmonic pulses. Nonlinear cascaded method will provide a reference for the efficient frequency conversion in the high intensity range.

Keywords: Femtosecond; Third Harmonic Generation; Second Harmonic Generation; Cascaded Nonlinear Process

1. Introduction

Frequency upconversion is a useful nonlinear process to achieve short wavelength, and third harmonic generation (THG) is one of the most effective upconversion process, which can be used to generate optical emission with shorter wavelength. Generally, third-order susceptibility of material contributes to THG process, i. e., direct THG. Besides direct THG method, cascaded THG by second harmonic and sum frequency process is a more efficient method. Higher conversion efficiency can be achieved with quadratic nonlinearity using two cascaded nonlinear processes.

However, the most efficient conversion of cascaded THG is achieved when the phase-matching conditions for both second harmonic and sum frequency processes are satisfied. In case of cw or quasi-cw regimes, the simultaneous phase-matching of the two parametric processes has been realized in several different structures in the past two decades [1-3]. For two-dimension nonlinear photonic crystals, they are often utilized to realize the noncollinear THG [4-6]. Recently, collinear THG was also achieved by a short-range-ordered two-dimension nonlinear photonic crystal [7]. However, there are few reports on cascaded THG of ultrashort pulses. N. Fujioka realized noncollinear cascaded THG of femtosecond pulses using two-dimension periodically poled lithium niobate and THG efficiency of 8% with spectral width of 4 nm was obtained [8].

In this paper, we demonstrated a collinear cascaded THG of femtosecond pulses with high intensity in a 5 mol. % MgO-doped periodically poled lithium niobate crystal. In the single pass scheme, the optimal THG efficiency of 10.8% was obtained at the input intensity of 74 GW/cm^2. The TH spectral width reached as broad as 8.7 nm with a small group velocity mismatch (GVM) between fundamental pulses and SH pulses.

2. Experimental Configuration

In general, a periodic QPM material can provide only one effective wave vector for a parametric process. However, a QPM material with a period of Λ is also possible to provide two effective wave vectors contributing to the two QPM processes, respectively. In such a periodic QPM marital, the cascaded THG, which involves the simultaneous SHG and SFG processes, with the respective Δk_I and Δk_{II} as follows:

$$\begin{aligned}\Delta k_I &= k_2 - 2k_1 - k_{QPM1} \\ &= 4\pi(n_2 - n_1)/\lambda_1 - m_1 2\pi/\Lambda \\ \Delta k_{II} &= k_3 - k_2 - k_1 - k_{QPM2} \\ &= 2\pi(3n_3 - 2n_2 - n_1)/\lambda_1 - m_2 2\pi/\Lambda\end{aligned} \quad (1)$$

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where the subscripts 1, 2, and 3 represent the fundamental, SH and TH waves, respectively. k_i and n_i are the wave vector and the refractive index for the i-th wave (I = 1, 2, 3), k_{QPM1} and k_{QPM2} are the QPM wave vectors of m_1 and m_2 orders for SHG and SFG, respectively.

From the Sellmeier equations of 5 mol. % MgO-doped congruent $LiNbO_3$ [9], it is found that optimal grating period of 20.37 μm can be used to achieve an efficient cascaded THG under 35℃, provided that the polarization of the fundamental wave is chosen to be ordinary, and the polarizations of the SH and TH waves are chosen to be extraordinary. For the femtosecond pulses with a large spectral width, it has a large phase-matching bandwidth in the frequency conversion [10]. Therefore, the efficient SFG can still be achieved with the existence of a small wave-vector mismatch.

Considering the refractive index changes induced by the nonlinearity caused by the high intensity [11], the QPM conditions may be slightly changed with respect to the above estimations. To overcome this problem, the MgO: PPLN sample in our experiment (made by HC Photonics Corporation) was composed of ten parallel periodically-poled structures with different periods around 20.37 μm (from 19.5 μm to 21.3 μm with the interval of 0.2 μm). The length and the thickness of the sample were 5 mm and 0.5 mm, respectively. Its two end surfaces were optically flat polished but uncoated. The polarizations of ordinary beam and extraordinary beam are chosen to be parallel to y-axis and parallel to z-axis, respectively. The x-axis was chosen as the propagation direction. The polarization of the ordinary fundamental wave was set to be parallel to y-axis.

The experimental schematic is shown in **Figure 1**. The fundamental light source was an optical parametric amplifier, pumped by regeneratively amplified Ti: sapphire laser, operated at a repetition rate of 1 kHz. The fundamental pulses at a central wavelength of 1550 nm had a pulse duration about 100 fs and a spectral width (FWHM) about 60 nm. A high-transmission mirror at 1550 nm behind the laser source was used to inhibit other wavelengths. A combination of a half-wave plate at 1550 nm and a Glen-Taylor prism was used to adjust the power of fundamental wave. When the Glen-Taylor prism is rotated to the right angle, the ordinary beam is allowed to pass only. A lens with a focal length of 200 mm is used to couple the fundamental beam into the sample. To avoid the crystal damage caused by the high intensity, we set the focus at a distance of 15 mm behind the output face of the crystal. The beam waist at the focus is about 50 μm. The radii of the beam in the input and output faces of the sample are 203.6 and 156.2 μm, respectively. The crystal was placed inside a temperature-controlled oven, in which the operation temperature can be controlled up to 200℃ with an accuracy of 0.1℃. Behind the oven, a focus lens, a high-reflecting mirror for the residual fundamental wave, and a band-pass filter for the SH or TH wave were used.

3. Results and Discussions

The dependences of the directly measured SH and TH power on the input power are shown in **Figure 2(a)**. Under our experimental condition, the input power of 10 mW corresponds to an input peak intensity of 100 GW/cm^2. The maximum input power is 54 mW, corresponding to an input peak intensity of 540 GW/cm^2. The highest SH efficiency of 4.5% is obtained at the input power of 13.3 mW (an input peak intensity of 133 GW/cm^2), while the highest TH efficiency of 10.8% is obtained at the input power of 7.4 mW (an input peak intensity of 74 GW/cm^2). When the losses are taken into account, composed of the coupling loss of 5% and the Fresnel losses (14.2% for the fundamental wave, 13.6% for the SH wave, 14.5% for the TH wave), the highest efficiencies of the generated SH and TH waves are 6.5% and 15.7%, respectively, as shown in **Figure 2(b)**. As the input power increases, the SH power increases linearly and the TH power increases quadratically. The THG efficiency saturation has been observed at the intensity level of 20 GW/cm^2 in the femtosecond cascaded THG [8]. When the intensity is raised to the order of magnitude of 100 GW/cm^2, the THG efficiency can't keep constant. It decreases with the increasing input intensity.

The spectra were measured by a high-resolution spectrometer (Ocean Optics), as shown in **Figure 3(a)**. The SH spectrum has a smooth profile with a FWHM of 4.7 nm. The main peak of the TH spectrum appears at 508.9 nm

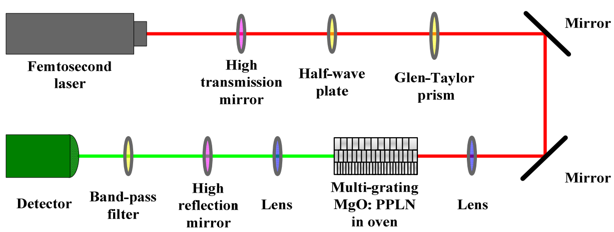

Figure 1. Experimental setup schematic.

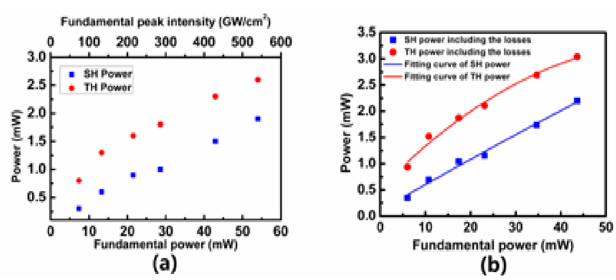

Figure 2. (a) Measured SH and TH power versus fundamental power; (b) SH and TH power including losses and fitting curves.

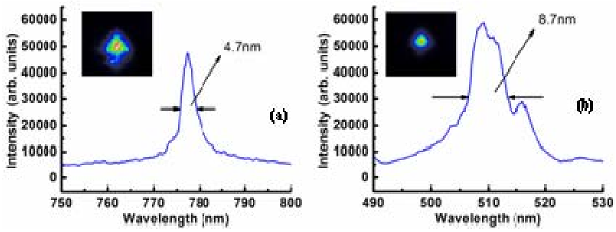

Figure 3. (a) SH spectra and mode; (b) TH spectra and mode.

with a FWHM of 8.7 nm and another peak locates at 516.1 nm. The TH spectral width is more than two times as the reported value of 4 nm by the cascaded THG of 118 fs pump pulses with a FWHM of 51 nm in the two-dimension PPLN [8]. The measured TH mode in the output face of the sample is shown in inset of **Figure 3(b)**. A nearly circular profile indicates a good spatial intensity distribution of THG.

During the cascaded processes, the TH bandwidth is limited by GVM among fundamental, SH and TH pulses. However, the GVM between the fundamental and SH pulses is the most important because it directly determines the effective interaction length of the SFG process [8]. In our experiment, the small GVM between the fundamental and SH pulses mainly results in the generation of the broadband TH wave. We choose the SHG type (o+o--e) that the fundamental and SH pulses have different polarizations. In the 5 mol. % MgO-doped congruent lithium niobate crystal, the GVM between ordinary fundamental wave (1550 nm) and extraordinary SH wave (775 nm) is only 155.6 fs/cm, while that is 3034 fs/cm for the SHG type (e+e--e) that the polarizations of the fundamental and SH pulses are both extraordinary [12].

4. Conclusions

Efficient cascaded femtosecond THG using a 5 mol. % MgO-doped multi-grating periodically poled crystal was experimentally demonstrated. The optimal TH efficiency of 10.8% was obtained in case of simultaneous phase matching of SHG and the SFG. The TH spectral width reached as abroad as 8.7 nm with a small GVM between the fundamental and SH pulses. Such nonlinear cascaded process will provide a reference for the efficient frequency conversion in the high intensity range.

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