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Coherent enhancement of broadband frequency up-conversion in BBO crystal by shaping femtosecond laser pulses

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Abstract

An optimal feedback control of broadband frequency up-conversion in BBO crystal is experimentally demonstrated by shaping femtosecond laser pulses based on genetic algorithm, and the frequency up-conversion efficiency can be enhanced by ~16%. SPIDER results show that the optimal laser pulses have shorter pulse-width with the little negative chirp than the original pulse with the little positive chirp. By modulating the fundamental spectral phase with periodic square distribution on SLM-256, the frequency up-conversion can be effectively controlled by the factor of about 17%. The experimental results indicate that the broadband frequency up-conversion efficiency is related to both of second harmonic generation (SHG) and sum frequency generation (SFG), where the former depends on the fundamental pulse intensity, and the latter depends on not only the fundamental pulse intensity but also the fundamental pulse spectral phase.

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Keywords: Coherent control; Frequency up-conversion; Second harmonic generation (SHG); Sum frequency generation (SFG); Pulse shaping

1. Introduction

Based on femtosecond pulse shaping techniques, coherent control can manipulate the laser-matter interaction by controlling quantum interference pathways, and thus desirable outcomes can be achieved [1–5]. As to the complicated quantum system, it is difficult to acquire the detail information of the quantum system, and the resulting laser field is often quite complicated and even impossible to be realized in the laboratory under the present technology. Recently, the successful emergence of the feedback-loop control technique develops an efficient method to optimize the laser field without any prior knowledge of the quantum system, and it has been widely applied on physical, chemical and biological processes [6–12].

Frequency up-conversion is one of the most useful nonlinear optical phenomena for extending the tunable wavelengths to shorter wavelengths [13–16]. When the pump waves with the frequency of ω_1 and ω_2 propagating in nonlinear crystal can generate a wave at $\omega_3 = \omega_1 + \omega_2$ due to nonlinear polarization $P^{(2)}$ ($\omega_3 = \omega_1 + \omega_2$). For effective energy transfer from the pump wave to the generated wave, both the energy and momentum conservation should be satisfied. The energy conservation requires $\omega_3 = \omega_1 + \omega_2$ and the momentum conservation requires $k_3 = k_1 + k_2$

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(phase-matching condition). That $\omega_1 = \omega_2 = \omega$ and $\omega_3 = 2\omega$ is known as second harmonic generation (SHG), and that $\omega_1 \neq \omega_2$ is known generally as sum frequency generation (SFG). When a femtosecond laser pulse propagates in the nonlinear crystal, due to the broadband spectral distribution, the frequency up-conversion signals may have complicated broadband spectra. The spectral phase structure of the fundamental laser determines the temporal order of the frequency components and should have a profound effect on the frequency up-conversion. In order to achieve high frequency up-conversion efficiency, it is very important to coherently manipulate the spectral amplitude and phase of the broadband fundamental laser pulses. In this paper, an optimal enhancement of broadband frequency up-conversion in 1 mm BBO crystal (Type I) is achieved experimentally by shaping femtosecond laser pulses based on genetic algorithm, and the frequency up-conversion can be effectively controlled by modulating the spectral phase with the periodic square distribution on SLM-256.

2. Experiment

The experimental setup for optimal feedback control is shown in Fig. 1. A Ti:sapphire mode-locked laser (Spitfire regenerative amplifier from Spectra-Physics Co.) is used as the excitation laser with the pulse duration of 50 fs, the repetition rate of 1 kHz and the center wavelength of 800 nm. The programmable 4-f pulse shaper is composed of a pair of diffraction gratings with 1200 lines/mm and a pair of spherical mirrors with a 150 mm focal length. A one-dimensional programmable liquid–crystal spatial light modulator array (SLM-256, CRI) is placed at the Fourier plane as an updatable filter for the femtosecond laser spectral manipulation. The shaped pulses, which phase and amplitude can be independently manipulated by SLM-256, are focused onto a 1 mm β-BBO crystal (Type I) with a lens of 20 mm focal length. The frequency up-conversion signal is detected by a photomultiplier and amplified by a lock-in. A computer is served for recording the data, evaluating the cost function, optimizing the spectral filter and updating SLM-256, and genetic algorithm is used as the optimal algorithm of feedback control. The original and optimal laser pulses are measured by SPIDER from APE Co.

3. Results and discussion

To demonstrate the optimal feedback control of frequency up-conversion in BBO crystal, genetic algorithm is used for global optimization of the spectral mask for the amplitude and phase of the laser pulses (as shown in Fig. 1). The spectral amplitude and phase of the shaped femtosecond laser pulses, manipulated by SLM-256, determine the frequency up-conversion intensity. Firstly, the initial voltage values are generated randomly and loaded on the pixels of SLM-256 as the first generation. The produced shaped femtoscond pulses are focused on a 1 mm β-BBO crystal and the corresponding frequency up-conversion signals will be generated and detected. Secondly, the fitness for the frequency up-conversion intensity is calculated and the corresponding cost value is obtained. Genetic algorithm allows a change in all of the pixels of the spectral mask and a decision whether the change is either accepted or rejected according to the calculated cost value. The voltage values of the new spectral mask for second generation can be attained by genetic algorithm operation (select, crossover and mutate). Their impacts on the frequency up-conversion intensity are determined and evaluated. So, the above-mentioned optimization procedure is repetitively

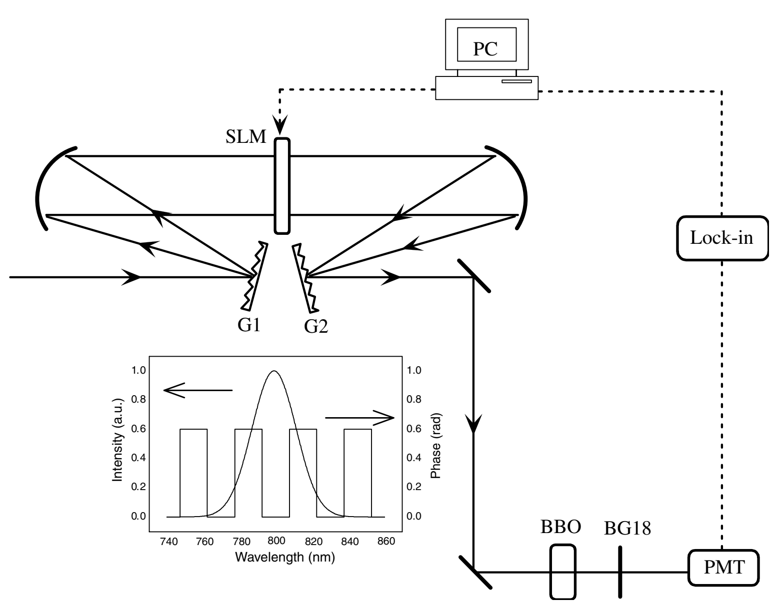

Fig. 1. Experimental setup for the coherent enhancement of frequency up-conversation.

performed till the cost value approaches convergence and the frequency up-conversion signals approach to the optimal maximum.

Fig. 2a shows the dependence of the broadband frequency up-conversion intensity on the iteration number. The broadband frequency up-conversion intensity achieves the optimal maximum after 38 generations, and its intensity can be effectively enhanced by ∼16%. The pulse-width and spectral phase of the original and optimal laser pulses are measured by SPIDER (as shown in Fig. 2b and c). The experimental results show that the optimal pulse can be compressed from 69 fs of the original pulse to 51 fs, and the optimal pulse has been optimized from the positive chirp to the negative chirp with less distribution.

As to well known, the evolution of the frequency up-conversion envelope function E_s can be expressed by,

$$E_u(\omega_u) = \int_{-\infty}^{\infty} \Gamma(z)\exp(\mathrm{i}\ k_u(\omega_u)z)\mathrm{d}z \times \int_{-\infty}^{\infty} E_f(\omega_f)E_f(\omega_u-\omega_f)\exp(-\mathrm{i}[k_f(\omega_f) + k_f(\omega_u-\omega_f)]z)\mathrm{d}\omega_f \quad (1)$$

where $\Gamma(z)$ is the nonlinear coupling coefficient, and E_f are the Fourier transform of the field envelope function, and k_f and k_u are the wave vectors of the fundamental wave and the up-conversion wave, respectively. For the broadband femtosecond pulse, the frequency up-conversion depends on not only the fundamental pulse intensity but also the fundamental pulse spectral phase. To investigate the dependence of frequency up-conversion in BBO crystal on the spectral phase, we modified the phases of the fundamental spectral components by applying spectral phase-only filters with the periodic square distribution to SLM-256,

$$\Phi(\omega) = \frac{\alpha}{2} + \frac{2\alpha}{\pi}\sum_{l=0}^{\infty}\frac{\sin[(2l+1)(\beta\omega+\phi)]}{2l+1} \quad (2)$$

where α and β are the modulation depth and modulation frequency, respectively, and ϕ is the modulation phase (as shown in Fig. 1, inset, for a typical phase modulation). Note that a phase-only modulation has no effects on the pulse energy or power spectrum.

Fig. 3 shows frequency up-conversion signal as a function of the modulation depth, modulation frequency and modulation phase, respectively. The experimental results accord with the theoretical fitting curve. The strong dependences of the frequency up-conversion signal on the modulation depth α and modulation frequency β and modulation phase ϕ are observed. As shown in Fig. 3a, as the modulation depth α increasing, the frequency up-conversion intensity is periodically modulated and reproduced as that for the transform-limited pulses at $\alpha = m\pi$ ($m = 1, 2, 3,\ldots$). The reason is that the periodically modulated shaped pulses can be reconstructed into the transform-limited pulses at $\alpha = m\pi$ ($m = 1, 2, 3, \ldots$) as the modulation depth α increasing. As shown in Fig. 3b, as the modulation frequency β increasing, the frequency up-conversion intensity shows a decay with a concomitant oscillation. As shown in Fig. 3c, the frequency up-conversion intensity as the spectral phase distribution is continuously transformed from symmetric to antisymmetric. It is noted that simply shifting

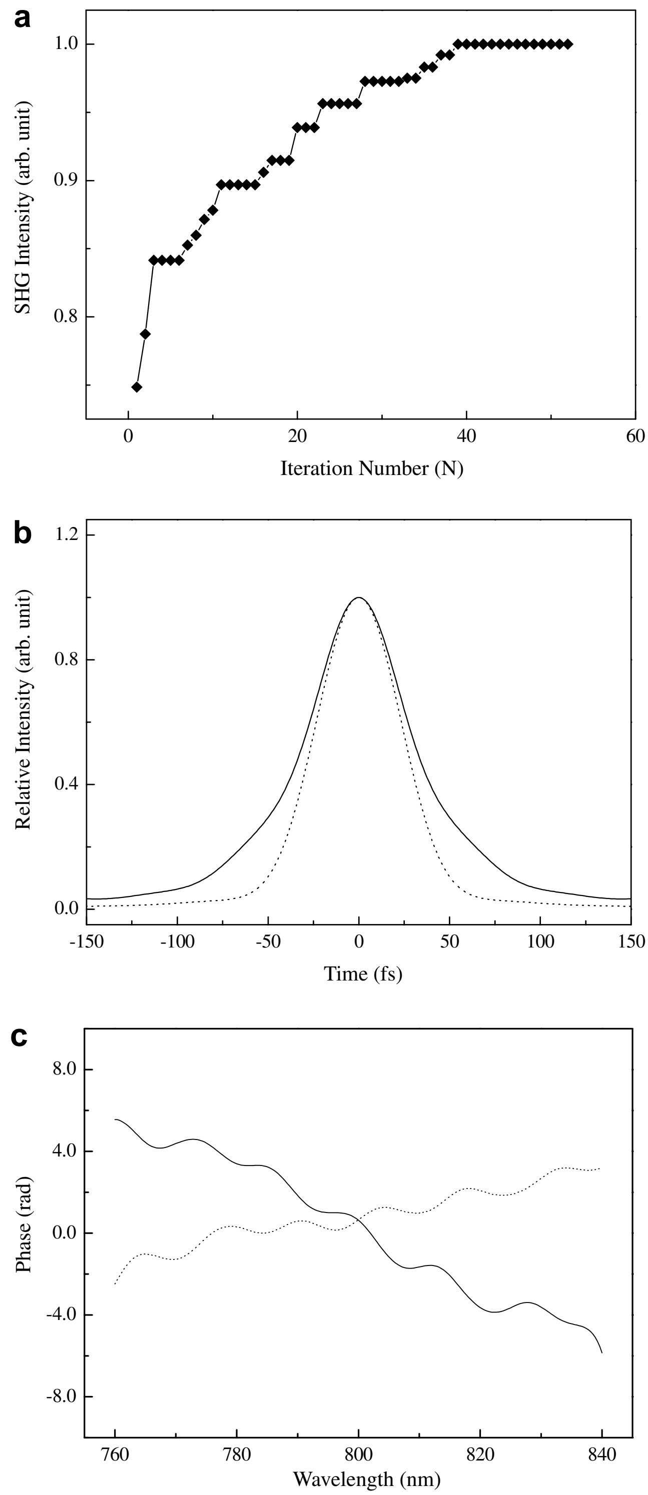

Fig. 2. (a) Optimal feedback up-conversation signal as a function of the iteration number based on genetic algorithm. The normalized pulse-width (b) and the spectral phase (c) for the original (solid line) and optimal (dash line) pulses from SPIDER, respectively.

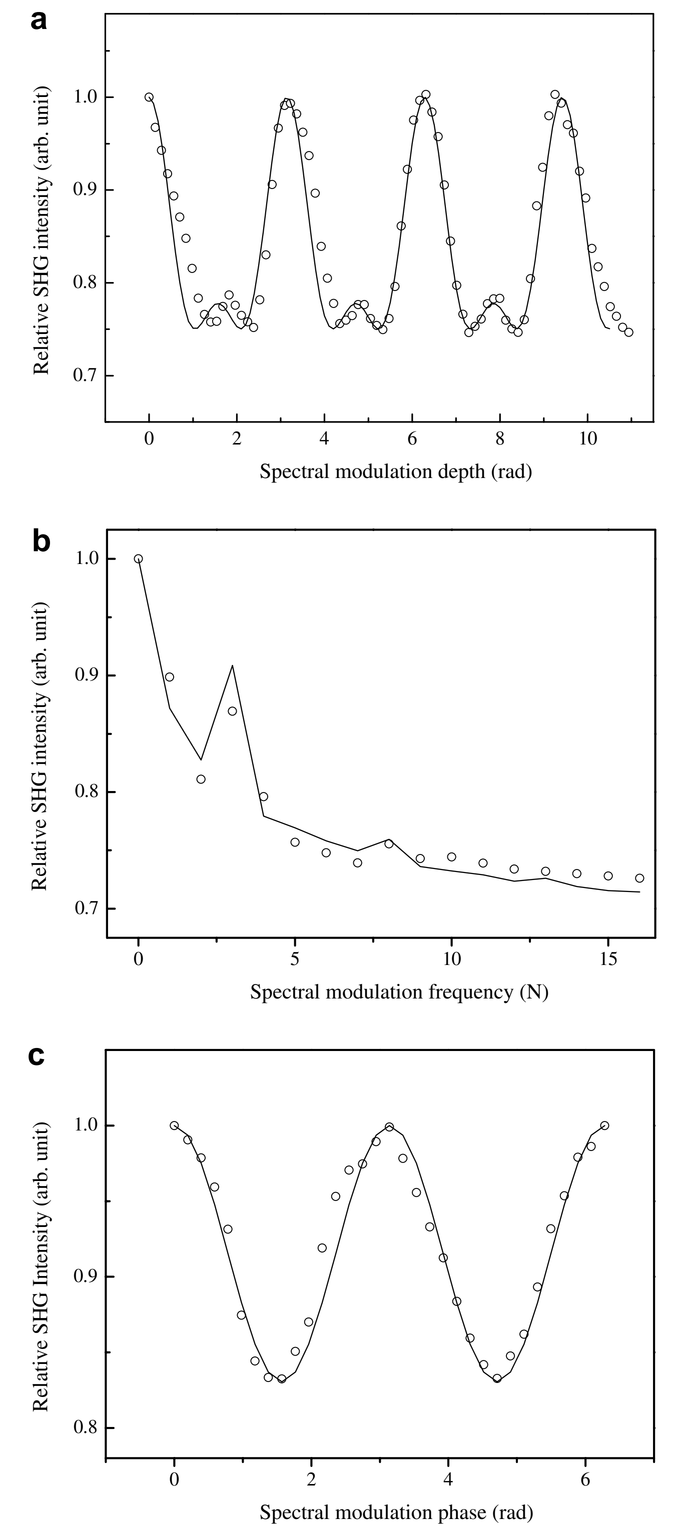

Fig. 3. Experimental (circle) and calculated (line) up-conversation signal as the function of the spectral modulation depth α for $\beta = 8$ and $\varphi = 0$ (a), and the spectral modulation frequency β for $\alpha = 8$ and $\varphi = 0$ (b) and the spectral modulation phase φ for $\alpha = 8$ and $\beta = 4$ (c), respectively.

the modulation phase across the pulse spectral affect neither the total power nor the power spectrum of the pulse. However, the frequency up-conversion intensity is modulated by the factor of about 17% (as shown in Fig. 3c.)

For the femtosecond laser pulse, the frequency up-conversion in BBO crystal may include second harmonic generation (SHG) from the same fundamental frequency components and sum frequency generation (SFG) from the different fundamental frequency components. Second harmonic generation (SHG) depends only on the fundamental pulse intensity, and so second harmonic generation (SHG) can be effectively enhanced by the pulse compression. However, sum frequency generation (SFG) depends on both of the fundamental pulse intensity and the fundamental spectral phase, and it is maximized by the transform-limited pulse ($\Phi(\omega_f) = \Phi(\omega_u - \omega_f) = 0$) and the anti-symmetric spectral phase distribution ($\Phi(\omega_f) = -\Phi(\omega_u - \omega_f) \neq 0$). So, the frequency up-conversion efficiency is related to both of the fundamental spectral phase independent SHG and the fundamental spectral phase dependent SFG, and it can be coherently controlled by modulating the effectiveness of the fundamental pulse spectral phase on the SFG components.

When the fundamental femtosecond pulse propagates in BBO crystal, because of the positive dispersion of BBO crystal, there is the group velocity dispersion of the fundamental frequency components, and it results in the fundamental pulse broadening and the fundamental spectral phase mismatching. So, the optimal negative chirp can compensate both of the fundamental pulse broadening and the fundamental spectral phase mismatching. On the one hand, the optimal compensation for the fundamental pulse broadening may result in the pulse compression and increase the pulse intensity-dependent second harmonic generation (SHG) and sum frequency generation (SFG). On the other hand, the optimal compensation for the fundamental spectral phase mismatching may improve the phase-matching condition the fundamental spectral phase dependent sum frequency generation (SFG).

4. Conclusion

In conclusion, the broadband frequency up-conversion generation in BBO crystal has been coherently enhanced ~16% by shaping femtosecond laser pulses based on genetic algorithm. The optimal laser pulses have shorter pulse-width with the little negative chirp than the original pulse with the little positive chirp. By modulating the fundamental spectral phase with periodic square distribution on SLM-256, the frequency up-conversion can be effectively controlled by the factor of about 17%. The experimental results indicate that the frequency up-conversion efficiency is related to both of the pulse intensity dependent second harmonic generation (SHG) and the pulse spectral phase dependent sum frequency generation (SFG). The optimal pulse compression can improve the conversion efficiency of second harmonic generation (SHG) and sum frequency generation (SFG), and sum frequency generation (SFG) can be further enhanced by the coherent optimization of the fundamental pulse spectral phase. So, the broadband frequency up-conversion can be efficiently enhanced by optimizing the fundamental pulse intensity and spectral phase.

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