Study of narrow-band second harmonic generation from a broad-band fundamental pulse*

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This paper studies the type-I phase-matched second harmonic generation using 25-fs input laser pulses in a thick BBO crystal. The harmonic signal exhibits a narrow spectrum bandwidth, even though the input pulse has a broad bandwidth. The energy transfer efficiency and modulation of the fundamental spectrum are investigated.

Keywords: second harmonic generation, cascaded process, energy transfer, chirp

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1. Introduction

One of the most typical quadratic nonlinear optical processes is second harmonic generation (SHG). Improving the conversion efficiency and acceptance bandwidth in SHG has attracted considerable attention in recent years.[1−5] An interesting phenomenon during SHG is that the fundamental spectrum can be significantly affected. This fundamental modulation is a Kerr-like effect and can be explained through cascaded quadratic nonlinearities:[6,7] In type-I phase-mismatched SHG, the fundamental pulses experience a phase shift caused by cascaded second-order processes when the phase-matching is not well satisfied ($\Delta k \neq 0$).[8,9] In type-II phase-matched SHG, there is a similar phase shift when the phase-matching condition is satisfied with a pre-delay of input ordinary and extraordinary fundamentals. Group-velocity mismatching has also been shown to be very important.[10,11] However, cascaded processes under type-I phase-matching conditions have not yet been discussed in depth.

The effect of two second-order nonlinear polarizations on the fundamental spectrum is similar to one third order nonlinear polarization: they all induce phase modulation. This phase modulation leads to spectral broadening of the fundamental pulses.[12−16] Because the Kerr-like effect induced by cascaded processes is based on phase modulation, it is interesting to determine how the output fundamental spectrum is affected by introducing an initial chirp modulation.

In this article, we use 25-fs laser pulses with a broad bandwidth as the fundamental light. In previous studies, broad-band fundamental pulses were usually used to generate broad-band second harmonic (SH) pulses. In our experiment, type-I phase-matched narrow-band SH pulses are generated. We investigate the energy transfer from the broad-band fundamental light to the narrow-band SH signal, as well as the spectrum change of the fundamental light. It is found that the initial frequency chirp can modulate the SHG conversion efficiency and output fundamental spectrum significantly. The simulation result is consistent with the experimental result.

2. Experimental setup and results

In our experiment, laser pulses with durations of 25 fs are used as the fundamental light. The repetition rate of the laser is 3 kHz, and the highest energy is 800 μJ per pulse. The laser pulses propagate through a BBO crystal. The crystal (5 mm × 5 mm × 5 mm, cut at $\theta = 29°$, $\varphi = 0°$ for type-I SHG) is placed on a rotational stage T to control the incident angle. A diffuse reflection screen is placed just after the crystal to direct the light into a fibre spectroscopy device, which records the exit spectrum. The experimental setup is shown in Fig. 1.

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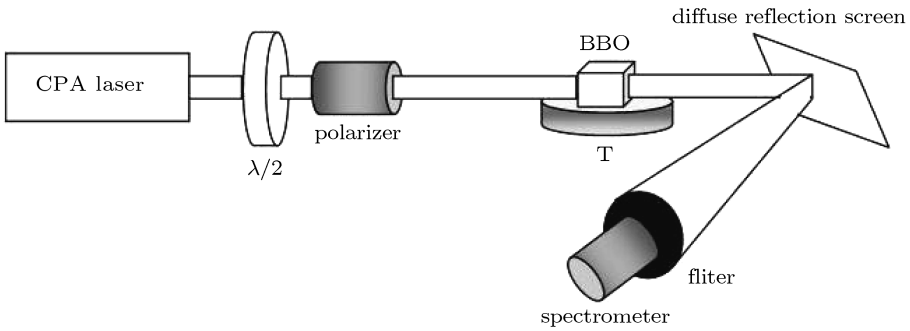


Fig. 1. Experimental setup.

The pump laser has a central wavelength of 791 nm, and the phase-matching wavelength of the BBO crystal can be adjusted by changing the incident angle. Figure 2(a) shows the contrast between the spectra of the pump laser and fundamental output signal when the wavelength of the SH signal is set to 395.85 nm. Figure 2(b) shows the corresponding spectrum of the SH pulses. We can see that the fundamental output pulse experienced modulation over a wide range. Wavelengths around the phase-matching point experienced energy loss while others experienced a gain. The full width at half maximum (FWHM) bandwidth of the main fundamental energy reduction is found to be 42 nm and that of the SH spectrum is 0.7 nm. It is obvious that the energy reduction of the fundamental signal occurs over a much broader range than the gain in the SH signal. This means that the broad-band fundamental modulation can induce a broad-band energy transfer to the SH signal. We found that 87.8% of the energy reduction from 763 nm to 832 nm is transferred to the SH signal, and the remaining 12.2% is transferred to the edge wavelengths of the fundamental pulse. The energy transfer efficiency from the input pulse to SHG is measured as 25%. We also find the ratio of energy in the SH signal from 394.5 nm to 397.5 nm to that in the initial fundamental signal from 789 nm to 795 nm to be 2.3. This result implies an essential effect of this energy transfer process to increase the SH/fundamental ratio.

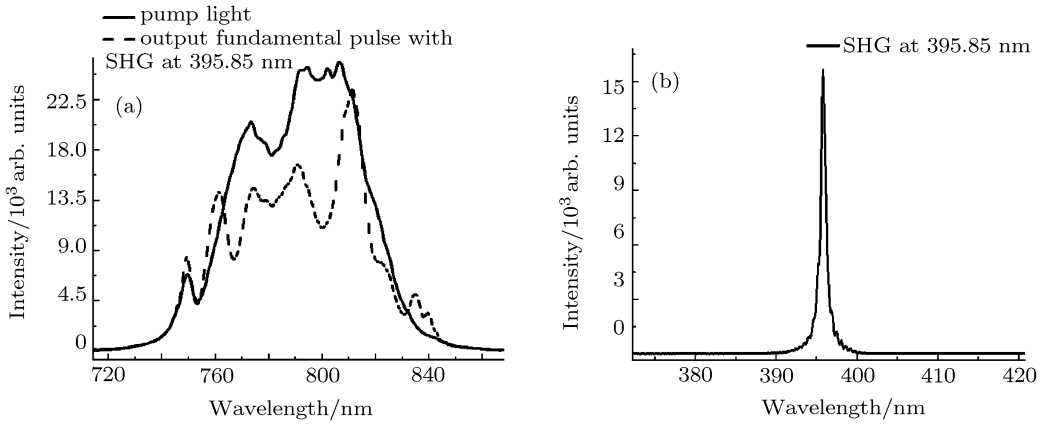


Fig. 2. (a) Contrast between the spectra of the pump laser (solid line) and output fundamental signal with SHG at 395.85 nm (dashed line); (b) SH spectrum at 395.85 nm.

When we change the wavelength of the phase-matching SHG by adjusting the incident angle, the SHG efficiency also changes. However, the process of energy transfer from the broad-band fundamental spectrum to narrow-band SHG and from middle wavelengths to the edge wavelengths remains unchanged. Figure 3 shows the exit spectra at different phase-matching wavelengths. The intensity decreased area and increased area are shifted with the shift of the phase-matching wavelength.

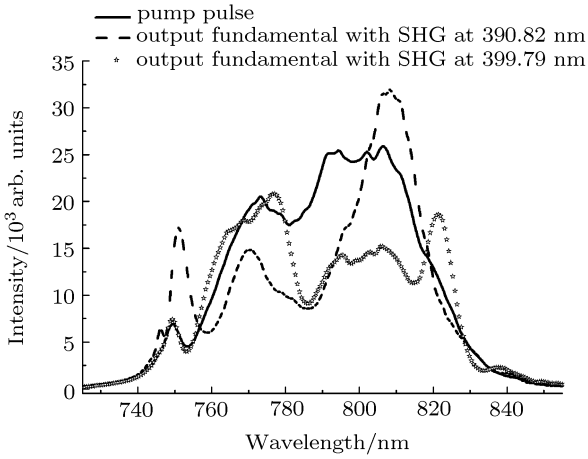


Fig. 3. Output fundamental spectra with different phase-matching wavelengths.

As is well known, ultra-short laser pulses experience self-phase modulation during propagation due to the Kerr effect. The pulses can also be modulated by cascaded second-order nonlinear polarization during phase-mismatched type-I SHG or phase-matched type-II SHG, where a portion of the energy is transferred from middle to edge wavelengths.[13,14] Similar effects were observed in our experiment. We can see that the wavelength range does not broaden in our experiment, while the Kerr effect would lead to obvious wavelength range broadening. Therefore, we postulate that the fundamental modulation is caused by only the cascaded second-order nonlinear processes.

3. Numerical simulation

To understand this broad-band fundamental modulation we simulated the propagation of a femtosecond laser pulse in a BBO crystal. For simplicity, we used the plane-wave approximation. The electric fields of the pulses are expressed as:

$$E_1(z,t) = \frac{1}{2}A_1(z,t)\exp(-\mathrm{i}\,\omega_1 t + \mathrm{i}\,k_1 z) + \text{c.c.}, \quad (1a)$$

$$E_2(z,t) = \frac{1}{2}A_2(z,t)\exp(-\mathrm{i}\,\omega_2 t + \mathrm{i}\,k_2 z) + \text{c.c.}, \quad (1b)$$

where $\omega_2 = 2\omega_1$, $k_1 = (\omega_1 \times n_\mathrm{o}(\omega_1))/c$, and $k_2 = (\omega_2 \times n_\mathrm{e}(\omega_2))/c$ are the central frequencies and corresponding wave-vectors of the SH and fundamental pulses, respectively, and the subscripts 1 and 2 indicate the fundamental and SH pulses, respectively. We can then apply the slowly-varying amplitude approximation[17,18] to Maxwell's equations to simulate SHG. The equations for the coupled fundamental and SH pulses propagating in the moving frame $\tau = t - k'(\omega_0)z$ are given by

$$\frac{\partial A_1}{\partial z} + (k_1' - k_2')\frac{\partial A_1}{\partial t} + 0.5\,\mathrm{i}\,k_1''\frac{\partial^2 A_1}{\partial t^2} = \mathrm{i}\,\frac{\omega_1\chi^{(2)}}{2n_1 c}A_2 A_1^* \exp(\mathrm{i}\,\Delta k z), \quad (2a)$$

$$\frac{\partial A_2}{\partial z} + 0.5\,\mathrm{i}\,k_2''\frac{\partial^2 A_2}{\partial t^2} = \mathrm{i}\,\frac{\omega_2\chi^{(2)}}{2n_2 c}A_1^2 \exp(-\mathrm{i}\,\Delta k z), \quad (2b)$$

where $\Delta k = k_2 - 2k_1$ is the wave vector mismatch. The central frequency of the fundamental and SH pulses are ω_1 (corresponding to 800 nm) and ω_2 (corresponding to 400 nm), respectively.

The initial pump pulse is set as

$$\begin{aligned} A_1(0,t) = {} & A_0 \exp\left[-\mathrm{i}(\omega_0-\omega_1)t - \frac{t^2}{(2.5\times t_\mathrm{p})^2}\right] + 0.15A_0\exp\left[-\mathrm{i}(\omega_3-\omega_1)t - \frac{t^2}{(6.5\times t_\mathrm{p})^2}\right] \\ & + 0.3A_0\exp\left[-\mathrm{i}(\omega_4-\omega_1)t - \frac{t^2}{(5\times t_\mathrm{p})^2}\right] + 0.45A_0\exp\left[-\mathrm{i}(\omega_5-\omega_1)t - \frac{t^2}{(4\times t_\mathrm{p})^2}\right], \end{aligned}$$

where $t_\mathrm{p} = 21.63$ fs and ω_0, ω_3, ω_4, ω_5 correspond to 791, 765, 775, and 806 nm, respectively, in order to fit the experimental input pulses. The A_0 is set as 0.6175×10^8 V/m (corresponding to an intensity of 0.5×10^{11} W/cm^2) and the propagation distance is 4.5 mm. The BBO refractive index is calculated by using the Sellmeier equations[19]

$$n_\mathrm{o}^2 = A_{\mathrm{o}1} + \frac{A_{\mathrm{o}2}}{\lambda^2 - A_{\mathrm{o}3}} - A_{\mathrm{o}4}\lambda^2, \quad n_\mathrm{e}^2 = A_{\mathrm{e}1} + \frac{A_{\mathrm{e}2}}{\lambda^2 - A_{\mathrm{e}3}} - A_{\mathrm{e}4}\lambda^2, \quad (\lambda \text{ in micrometers})$$

$$A_{\mathrm{o}1} = 2.7405, \quad A_{\mathrm{o}2} = 0.1839, \quad A_{\mathrm{o}3} = 0.0179, \quad A_{\mathrm{o}4} = 0.0155,$$

$$A_{\mathrm{e}1} = 2.373, \quad A_{\mathrm{e}2} = 0.156, \quad A_{\mathrm{e}3} = 0.044, \quad A_{\mathrm{e}4} = 0.027405.$$

The second-order nonlinear optical coefficient is set to $\chi^{(2)} = 2 \times 10^{-12}$ m/V.

The simulated spectrograms of the fundamental pulses are shown in Fig. 4. We can see from the simulation spectrum that the fundamental output pulse is modulated and the energy reduction bandwidth of the fundamental signal is much broader than that of SH signal. In our simulation, only second-order nonlinear polarization is considered, so the spectrum change is caused by $P(2\omega)$ and $P(\omega)$. The $P(2\omega)$ includes SHG and sum frequency generation (SFG) and $P(\omega)$ includes difference frequency generation. These two cascaded second-order nonlinear polarizations couple energy at different wavelengths of the fundamental and harmonic signals and induce the transfer of energy between different wavelengths. For example, figure 5 shows all the phase-matching SFG wavelengths at the incident angle for type-I phase-matching SHG at 400 nm. Fundamental wavelengths from 760 nm to 850 nm contribute to phase-matched SFG from 399.5 nm to 400 nm. This explains the broad-band range of energy reduction in the fundamental signal and the narrow-band range of gain in the harmonic signal.

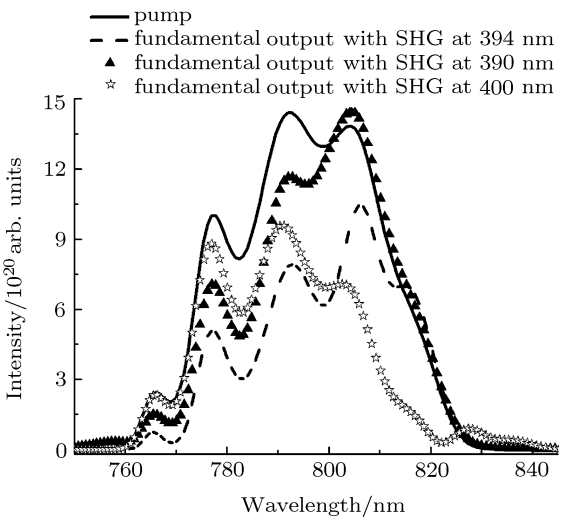


Fig. 4. Simulation results for contrast between the spectra of the pump laser pulse (solid line) and the fundamental output pulse with SHG at 390 nm (black triangle), 394 nm (dashed line) and 400 nm (star).

It is worth while pointing out that the modulation of the fundamental spectrum in the simulated results is smaller than the experimental results. This is because the pump laser in our experiment is not Fourier-transform limited, i.e., there is some chirping in the input pulse. The pulse duration is already minimized by adjusting the compressed coupling prism of the laser, so the chirp is of a high order.

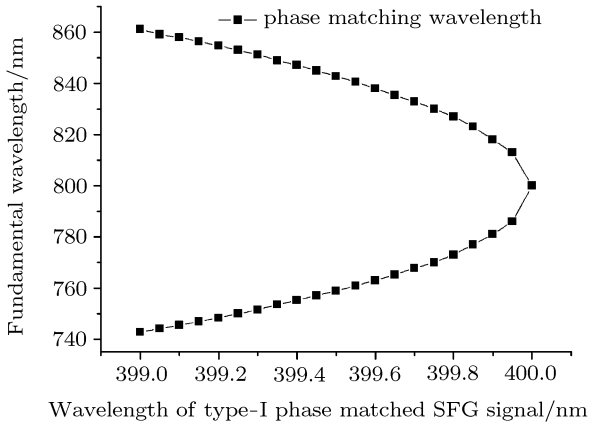


Fig. 5. Phase-matched SFG wavelength.

To determine how the chirp influences the fundamental output spectrum, we compared numerical results with different chirps as shown in Fig. 6. In this simulation, we added an initial phase modulation $A_1'(0,t) = A_1(0,t)\exp[\pm \mathrm{i} \cdot \pi \cdot k(\omega - \omega_1)^2]$, where $k = 0.86 \times 10^{-28}$.

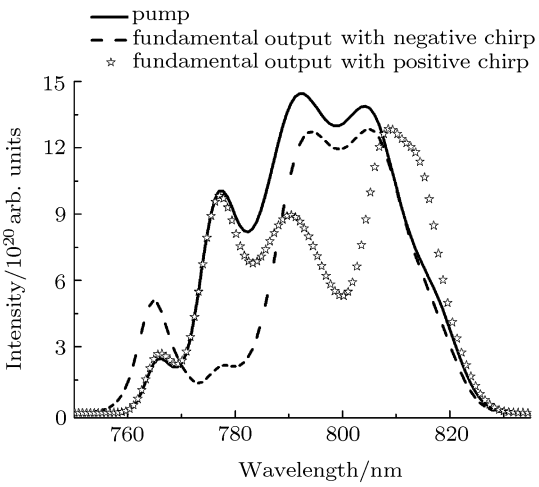


Fig. 6. Fundamental output spectrum with initial chirps.

The result shows that both the fundamental spectrum and the harmonic conversion efficiency are affected by initial chirp. In Fig. 6, the corresponding SHG efficiencies are 12.7% (with negative chirp), 25.15% (with positive chirp) and 43.94% (without chirp) respectively. The conversion efficiency change is due to the decrease of the fundamental peak intensity, which is caused by the chirp-induced duration broadening. From Fig. 6 we also see that initial chirp

would enhance modulation in the output fundamental spectrum and that positive and negative chirps lead to an increase in longer and shorter wavelengths, respectively. It is reasonable that the high-order chirp in our experiments causes both the shorter and longer wavelengths to experience enhanced modulation.

After including the initial chirp in our simulation, the results match the experiment better. Both the experimental and numerical results prove that the initial chirp can modulate the fundamental pulse and the harmonic conversion efficiency significantly.

4. Conclusions

In conclusion, we find that when a broad-band fundamental pulse is used to generate narrow-band phase-matched SHG, the fundamental pulse experiences a broad-band modulation, which transfers energy from the broad-band fundamental to the narrow-band SH signal. In addition, we find that the initial laser chirp can modulate the SHG conversion efficiency and the fundamental output spectrum significantly.

References

[1] Fejer M M, Magel G A, Jundt D H and Byer R L 1992 *IEEE J. Quantum Electron.* **28** 2631

[2] Coutts D W 1995 *IEEE J. Quantum Electron.* **31** 2208

[3] Steinbach A, Rauner M, Cruz F C and Bergquist J C 1996 *Opt. Commun.* **123** 207

[4] Shwa D, Eisenmann S, Marcus G and Zigler A 2009 *Opt. Express* **17** 6451

[5] Zhang H Z, Yang J B and Gao J Y 2003 *Chin. Phys.* **12** 518

[6] Huang G X 2001 *Chin. Phys.* **10** 418

[7] Desalvo R, Hagan D J, Sheikbahae M, Stegeman G, Vanstryland E W and Vanherzeele H 1992 *Opt. Lett.* **17** 28

[8] Xu G, Zhu H Y, Wang T and Qian L J 2002 *Opt. Commun.* **207** 347

[9] Su W H, Qian L J, Luo H, Fu X Q, Zhu H Y, Wang T, Backwitt K, Chen Y F and Wise F 2006 *J. Opt. Soc. Am.* B **23** 51

[10] Dubietis A, Valiulis G, Danielius R and Piskarskas A 1996 *Opt. Lett.* **21** 1262

[11] Dubietis A, Valiulis G, Danielius R and Piskarskas A 1998 *Pure Appl. Opt.* **7** 271

[12] Zeng X L, Ashihara S, Chen X F, Shimura T and Kuroda K 2008 *Opt. Commun.* **281** 4499

[13] Ashihara S, Shimura T, Kuroda K, Yu N E, Kurimura S, Kitamura K, Cha M and Taira 2004 *Appl. Phys. Lett.* **84** 1055

[14] Liu X, Qian L J and Frank Wise 1999 *Opt. Lett.* **24** 1777

[15] Xu G, Qian L J, Wang T, Zhu H Y, Zhu C S and Fan D Y 2004 *IEEE J. Sel. Top. Quantum Electron.* **10** 174

[16] Di Trapani P, Caironi D, Valiulis G, Dubietis A, Danielius R and Piskarskas A 1998 *Phys. Rev. Lett.* **81** 570

[17] Chien C Y, Korn G, Coe J S, Squier J and Mourou G 1995 *Opt. Lett.* **20** 353

[18] Krylov V, Rebane A, Kalintsev A G, Schwoerer H and Wild U P 1995 *Opt. Lett.* **20** 198

[19] Jundt D H, Fejer M M and Byer R L 1990 *IEEE J. Quantum Electron.* **26** 135