Generation of the cascaded fifth-order nonlinear phase shifts with femtosecond pulse

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Due to group velocity mismatch, the generation of the cascaded fifth-order nonlinear phase shifts with femtosecond pulse will inevitably cause the severe pulse distortion. By theoretical study and numerical simulations, we show that undistorted femtosecond pulse which is impressed the cascaded fifth-order nonlinear phase shifts can be obtained at large values of the phase mismatching ΔkL.

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The existence of nonlinear phase shifts produced by the cascaded third-order nonlinearity via $\chi^{(2)}:\chi^{(2)}$ cascading process on the fundamental wave was first discussed by Ostrovskii in 1967[1], and large cascaded third-order nonlinear phase shifts were measured in KTP[2,3] and periodically poled $LiNbO_3$ (PPLN)[4] later. Because of its large magnitude, the induced effective third-order nonlinearity from $\chi^{(2)}:\chi^{(2)}$ cascading can be as large as thousands times of Kerr nonlinearity[4], controllable sign and saturable feature, cascaded third-order nonlinear phase shifts have been found many important applications[5] such as spatio-temporal soliton generation[6], B-integral compensation[7], and the formation of all optical switching devices[8]. A configuration of phase-mismatched frequency tripling to induce effective fifth-order $\chi^{(5)}(\omega;\omega,\omega,\omega,-\omega,-\omega)$ nonlinearity exclusively via $\chi^{(2)}(\omega;3\omega,-2\omega):\chi^{(2)}(3\omega;2\omega,\omega):\chi^{(2)}(2\omega;\omega,\omega)$ cascading process has been proposed[9]. Compared with the cascaded third-order nonlinear phase shifts, the induced fifth-order nonlinear phase shifts can be five times larger in magnitude with a richer saturable feature. However on the femtosecond time scale, the generation of nonlinear phase shifts by the $\chi^{(2)}:\chi^{(2)}:\chi^{(2)}$ cascading process unavoidably meets group velocity mismatch (GVM) among the pulses. Because the fundamental pulse, its second harmonic and third harmonic have different group velocities, they move away from one another in time as propagating through the nonlinear crystal. This will severely distort the fundamental pulse while the large cascaded fifth-order nonlinear phase shifts are produced. Thus, the distortion of the fundamental pulse becomes an imposing obstacle to applications of the cascaded fifth-order nonlinear process. In this paper, we report an investigation of the nonlinear phase shifts accumulated by femtosecond-duration pulse undergoing the $\chi^{(2)}:\chi^{(2)}:\chi^{(2)}$ cascading process. We find that there is an advantage to working at relatively large phase mismatching ΔkL: distortion of the fundamental pulse due to GVM is weaker than that at small ΔkL. Finally under the adequately large ΔkL condition, undistorted fundamental pulse with induced fifth-order nonlinear phase shifts is achieved. In addition to mode-locking of lasers, the realization of large cascaded fifth-order nonlinear phase shifts with femtosecond pulse will be valuable to other applications such as pulse compression.

Based on the phase-mismatched frequency tripling, the cascaded fifth-order nonlinearity can be produced. We know that the third harmonic of laser beam is usually generated by $\chi^{(2)}(3\omega;2\omega,\omega):\chi^{(2)}(2\omega;\omega,\omega)$ cascading process: phase-matched second harmonic generation (SHG) followed by the phase-matched sum frequency generation (SFG). Obviously if the phase-matching condition can not be met in the second step of the frequency tripling, the phase-mismatched frequency mixing actually will contain two periodical processes, i.e. (i) SFG between the fundamental and its second harmonic, (ii) beating between the second harmonic and the third harmonic to generate a wave at the fundamental frequency. As a result, an additional phase retardation is impressed on the fundamental which is related to an effective $\chi^{(5)}$ nonlinearity. This can be expressed as $\chi^{(2)}(\omega;3\omega,-2\omega):\chi^{(2)}(3\omega;2\omega,\omega):\chi^{(2)}(2\omega;\omega,\omega)$ cascading, which is equivalent to the fifth-order nonlinearity $\chi^{(5)}(\omega;\omega,\omega,\omega,-\omega,-\omega)$ with degenerate frequencies.

The coupled wave-equations governing the fundamental, second harmonic and third harmonic in frequency mixing with the slowly varying envelope approximation are

$$\frac{\partial E_1}{\partial z}+\frac{1}{L_{\mathrm{GVM}_1}}\frac{\partial E_1}{\partial t}=-\frac{i}{2L_{\mathrm{NL}}}E_3E_2^*\exp(-i\Delta kz),\quad(1)$$

$$\frac{\partial E_2}{\partial z}=-\frac{i}{L_{\mathrm{NL}}}E_3E_1^*\exp(-i\Delta kz),\quad(2)$$

$$\frac{\partial E_3}{\partial z}+\frac{1}{L_{\mathrm{GVM}_2}}\frac{\partial E_3}{\partial t}=-\frac{3i}{2L_{\mathrm{NL}}}E_1E_2\exp(i\Delta kz),\quad(3)$$

where $L_{\mathrm{GVM}_1}=\tau/(1/v_2-1/v_1)$ is the temporal walk-off length between the fundamental and its second harmonic, $L_{\mathrm{GVM}_2}=\tau/(1/v_2-1/v_3)$ is the temporal walk-off length between the second harmonic and the third harmonic (with τ the second harmonic pulse duration, v_1 the fundamental pulse group velocity, v_2 the second harmonic group velocity and v_3 the third harmonic group velocity). Here the nonlinear length $L_{\rm NL} = 2n_\omega c/(\omega\chi^{(2)}E_1)$ is used to measure the input amplitude of the fundamental wave, and the phase mismatch is $\Delta k = k_{3\omega} - k_{2\omega} - k_\omega$. Under the conditions of low input intensity and negligible pump depletion, the cascaded fifth-order nonlinear phase shift impressed onto the fundamental wave at the exit surface of SFG crystal $z = L$ is given by[9]

$$\Delta\Phi^{\rm NL} = -\frac{3L_0^2L^2\omega^4|\chi^{(2)}|^4}{4c^6\varepsilon_0 n_\omega^3 n_{2\omega}^2 n_{3\omega}\Delta kL}I^2, \quad (4)$$

where L_0 is the length of SHG crystal. This nonlinear phase shift varies linearly with the square of input fundamental intensity I^2, and its sign is controlled by the phase mismatching ΔkL.

In cascaded fifth-order nonlinear process, GVM among the fundamental pulse, its second harmonic and third harmonic is difficult to avoid with femtosecond pulse-duration. Especially in the phase mismatched SFG crystal, the fundamental, second harmonic and third harmonic move with different group velocities. In order to quantify the effect caused by the GVM, the temporal walk-off length $L_{\rm GVM}$ is defined. There are two temporal walk-off lengths in phased mismatched SFG. One is $L_{\rm GVM_1}$ and the other is $L_{\rm GVM_2}$. Over these distances, the fundamental and the second harmonic or the second harmonic and the third harmonic will be separated by approximately one second harmonic pulse duration τ. Therefore the fundamental pulse produced by the difference frequency generation (DFG) between the third harmonic and the second harmonic will suffer from the severe distortion while the large cascaded fifth-order nonlinear phase shifts are impressed on it.

In fact, the generation of the cascaded fifth-order nonlinear phase shifts is related to one nonlinear cycle of up-conversion and down-conversion closely: firstly the fundamental and second harmonic via up-conversion generate the third harmonic; secondly owing to the phase mismatching, the second harmonic and the third harmonic beat each other to flow energy back into the fundamental pulse by down-conversion. If we can arrange for this cycle to occur before three pulses separate in time, for example in a distance shorter than $L_{\rm GVM_2}$ (assuming $L_{\rm GVM_1} > L_{\rm GVM_2}$), the undistorted fundamental pulse with the cascaded fifth-order nonlinear phase shifts can be realized. For a SFG crystal of length $L = NL_{\rm GVM_2}$, in order to avoid the deleterious effect of GVM at least N nonlinear cycles are required. According to this requirement the cascaded fifth-order nonlinear process must work at the large phase mismatching[10],

$$\Delta kL > 4N\pi. \quad (5)$$

When the phase mismatching ΔkL is large, the fundamental pulse experiences such nonlinear cycle several times as it traverses the SFG crystal. Each cycle of the up-conversion and down-conversion defines an effective interaction length that is shorter than the overall interaction, and the effects of GVM accumulated over the effective interaction length are therefore smaller than would be the case if the interaction length were the overall SFG crystal length. Distortion of the fundamental pulse should be less than at small phase mismatching ΔkL, where there are fewer nonlinear cycles and thus the effective interaction length is greater.

To describe the behaviors of the fundamental pulse with femtosecond-duration in a more general manner during the cascaded fifth-order nonlinear process, the coupled wave-equations have been solved numerically with the parameters of BBO crystal: input 100-fs fundamental wave $\lambda = 1$ μm, GVM between fundamental and the second harmonic $\rm GVM_1 = 100$ fs/mm (corresponding $L_{\rm GVM_1} = 0.7$ mm), GVM between the second harmonic and the third harmonic $\rm GVM_2 = 200$ fs/mm (corresponding $L_{\rm GVM_2} = 0.35$ mm). Numerical calculations (Fig. 1) indicate that with $\Delta kL = 0.1\pi$ after propagating through $L = 1.5$ mm SFG crystal, due to GVM the fundamental pulse has suffered from the distortion severely. The induced fifth-order nonlinear phase shifts on the fundamental nearly reach at π (Fig. 2). In this case the length of SFG crystal is almost five times as long as $L_{\rm GVM_2}$. So according to Eq. (5), for $L = 5L_{\rm GVM_2}$, the high-quality fundamental pulse can be generated only at the criterion $\Delta kL > 20\pi$ to be satisfied. The undistorted fundamental pulse with $\Delta kL = 28\pi$ which meets the criterion with ample margin is shown in Fig. 3. The trace of Fig. 3 explicitly demonstrates the advantage of using the cascaded process at large ΔkL: the pulse distortion caused by GVM is substantially reduced. In exchange for the improved pulse, the magnitude of the cascaded fifth-order nonlinear phase shifts decreases with ΔkL, but the decrease is naturally not so large if the cascaded nonlinear phase shifts are saturating at lower ΔkL. Even so, we also can further compensate this reduction by raising the SHG efficiency of the cascaded fifth-order nonlinear process. It is reasonable to conclude that at adequately large ΔkL, the cascaded fifth-order nonlinearity can be used for shaping femtosecond pulses as short as the walk-off time in the nonlinear medium[11].

The generation of large cascaded phase shifts naturally requires working under conditions of some saturation. The results of simulations show that GVM can lead to not only the pulse distortion but also the saturation of the cascaded fifth-order nonlinear phase shifts (Fig. 2). Over the length of SFG crystal $L = 0.5$ mm, the second

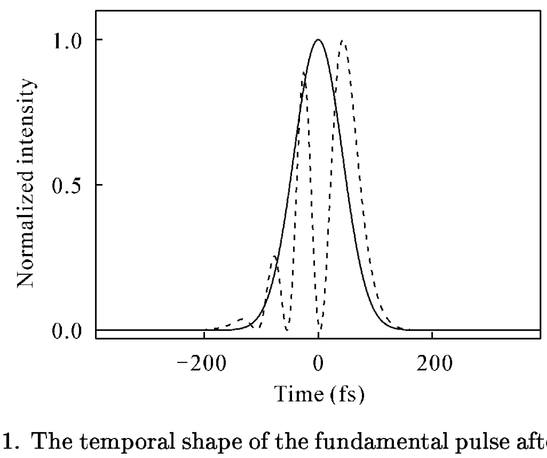

Fig. 1. The temporal shape of the fundamental pulse after the cascaded fifth-order nonlinear process with the parameters (dashed line): $L = 1.5$ mm, $L_{\rm NL} = 0.22$ mm, $\Delta kL = 0.1\pi$, $\rm GVM_1 = 100$ fs/mm, $\rm GVM_2 = 200$ fs/mm, and the efficiency of SHG 75% is used. The profile of initial fundamental with 100-fs pulse duration is also shown as a reference (solid line).

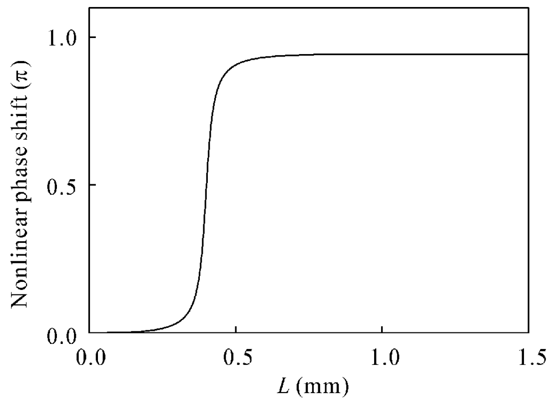

Fig. 2. The cascaded fifth-order nonlinear phase shifts of the fundamental pulse versus the SFG crystal length with the same parameters used in Fig. 1.

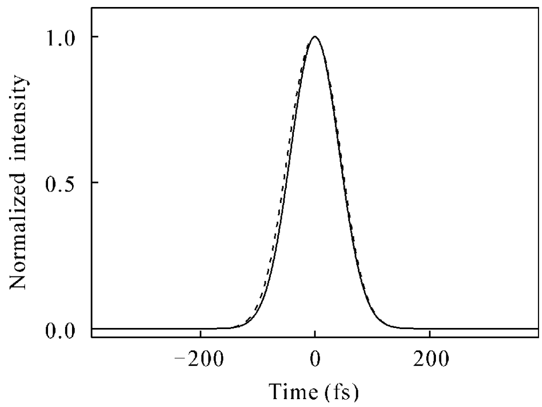

Fig. 3. The temporal shape of the fundamental pulse after the cascaded fifth-order nonlinear process for phase mismatching $\Delta kL = 28\pi$ (dashed line). Other parameters are the same as that in Fig. 1. The profile of initial fundamental with 100-fs pulse duration is also shown as a reference (solid line).

harmonic and the third harmonic have moved apart from each other entirely, because the temporal walk-off length between them is only L_{GVM_2}=0.35 mm. For this reason the fundamental pulse impressed nonlinear phase shifts can not be produced by DFG between the second harmonic and the third harmonic again. So with further propagation of pulses in the SFG crystal, the cascaded fifth-order nonlinear phase shifts almost do not show any increase but keep the magnitude of π constantly.

In conclusion, from the results of systematical study we find that the deleterious effects of GVM on the fundamental pluse with femtosecond pulse-duration can be eliminated by using the cascaded fifth-order nonlinear process at fairly large phase mismatch. Based on this, the undistorted fundamental pulse with cascaded fifth-order nonlinear phase shifts is obtained.

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