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# Nonlinear phase shift via multistep $\chi^{(2)}$ cascading

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**Abstract**

We present the first (according to our knowledge) consideration of the influence of the three- and four-step second-order nonlinearity cascading in the process of the nonlinear phase shift of the fundamental wave involved simultaneously on two nonlinear optical processes: second harmonic generation and sum frequency mixing. Under the condition that the two processes are nearly phase matched, the presence of multistep cascading leads to four times reduction of the input intensity necessary for obtaining a value of $\pi$ for the nonlinear phase shift. © 1998 Elsevier Science B.V. All rights reserved.

The effect of obtaining large nonlinear phase shift (NPS) by the fundamental wave participating in second-order nonlinear optical processes such as second harmonic generation (SHG) and sum frequency generation (SFG) via cascading was investigated extensively in the last few years [1–5].

The investigations show that the process of SFG or type II nondegenerate SHG is much more efficient if one would like to obtain larger NPS at lower input intensities [6–9]. In this case however the difficulty is that two light sources are needed and moreover only the weaker input wave obtains large NPS. On the other hand it is possible to realize simultaneously second harmonic generation and sum frequency mixing with only one fundamental input. The first study of simultaneous phase matching of SHG and SFM in ammonium oxalate crystal was presented in Refs. [10,11]. Simultaneous phase matching of three second-order processes in $LiNbO_3$ has been observed in Ref. [12]. Both works are based on birefringence phase matching. We should admit that simultaneous birefringence phase matching for two or three nonlinear processes is more or less accidental. With the quasi-phase-matching technique [13] it is now possible deliberately to construct nonlinear media that have simultaneous phase matching for more than one second-order process. This possibility has been demonstrated in KTP crystals [14] and $LiNbO_3$ crystals [15,16].

In this Letter we propose a method for obtaining large NPS with only one fundamental wave involved simultaneously in SHG and SFG.

Let us consider the fundamental beam with frequency $\omega$ entering a second-order nonlinear media. As a first step via a process of type I SHG the wave with frequency $2\omega$ is generated and as a second step via a process of SFG ($\omega + 2\omega = 3\omega$) a third harmonic is generated. Both processes (SHG and SFG) are supposed to be nearly phase matched. The generated second and third harmonic waves are downconverted to the fundamental wave $\omega$ via processes $(2\omega - \omega)$; $(3\omega - 2\omega)$ and $(3\omega - 2\omega,\ 2\omega - \omega)$ contributing to the nonlinear phase shift that the fundamental wave collects. As our calculations show this NPS that is the result of simultaneous action of two, three- and four-step $\chi^{(2)}$ cascading can exceed the value of $\pi$ for relatively low input intensities.

In the past three- and four-step $\chi^{(2)}$ processes have been considered only in connection with the possibility to generate a fourth harmonic in single noncentrosymmetric crystals with single input fundamental beam [17,18].

We start our investigations with the reduced amplitude equations in the slowly-varying envelope approximation,

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with the assumption of zero absorption for all interacting waves, that have the following form:

$$\frac{\mathrm{d}A_1}{\mathrm{d}z} = -\mathrm{i}\sigma_1 A_3 A_2^* \exp(-\mathrm{i}\Delta k_3 z) - \mathrm{i}\sigma_2 A_2 A_1^* \exp(-\mathrm{i}\Delta k_2 z), \quad (1a)$$

$$\frac{\mathrm{d}A_2}{\mathrm{d}z} = -\mathrm{i}\sigma_4 A_3 A_1^* \exp(-\mathrm{i}\Delta k_3 z) - \mathrm{i}\sigma_5 A_1 A_1 \exp(\mathrm{i}\Delta k_2 z), \quad (1b)$$

$$\frac{\mathrm{d}A_3}{\mathrm{d}z} = -\mathrm{i}\sigma_3 A_1 A_2 \exp(\mathrm{i}\Delta kz), \quad (1c)$$

where the subscripts '1' denote the fundamental waves, subscripts '2' denote the second harmonic and the subscript '3' denotes the third harmonic wave. The wave vector mismatch for the SHG process is $\Delta k_2 = 2k_2 - k_1$ and for the SFM process is $\Delta k_3 = k_3 - k_2 - k_1$.

As follows from system (1) there are three possible channels for phase modulation of the fundamental wave (see Fig. 1). The first channel is two-step cascading and has the same behaviour as in the case of type I SHG. The second channel represents three-step cascading and the third channel four-step cascading. The presence of the latter two channels leads to significant change of the nonlinear phase shift collected by the fundamental wave in comparison with the case of simple type I SHG.

The exact solution of system (1) must be obtained by numerical simulations, however using the negligible pump depletion approximation it is possible to obtain an analytical expression for the nonlinear phase shift due to multistep cascading. In this type of approximation [19,20] constant intensity of the fundamental wave, $|A_1|^2 = a_1^2 = a_1^2(0)$, but possible changes of the phase $\varphi_1$ of the fundamental wave are suggested. This approximation is valid for low input intensities ($\sigma a_1 \ll \Delta k$).

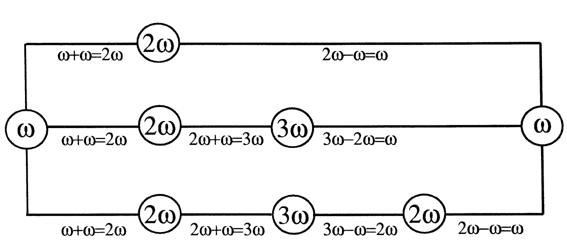


Fig. 1. Schematic drawing of possible channels for phase modulation of the fundamental wave.

Eq. (1a) can be split into the following equations for the real amplitude $a_1$ and the phase $\varphi_1$ of the fundamental wave ($A_1 = a_1 \exp(\mathrm{i}\varphi_1)$),

$$\frac{\mathrm{d}a_1}{\mathrm{d}z} = \sigma_1 a_3 a_2 \sin(\Phi_3) + \sigma_2 a_2 a_1 \sin(\Phi_2), \quad (2)$$

$$\frac{\mathrm{d}\varphi_1}{\mathrm{d}z} = -\sigma_1 \frac{a_3 a_2}{a_1} \cos(\Phi_3) - \sigma_2 a_2 \cos(\Phi_2). \quad (3)$$

In Eqs. (2) and (3)

$$\Phi_3 = \varphi_3 - \varphi_2 - \varphi_1 - \Delta k_3 z,$$
$$\Phi_2 = \varphi_2 - 2\varphi_1 - \Delta k_2 z \quad (4)$$

are introduced.

When the condition $\sigma a_1 \ll \Delta k$ is fulfilled, the fundamental amplitude is weak enough so system (1) can be solved by using a perturbation expansion of $\varphi_1$,

$$\varphi_1 = \varphi_{10} + \Delta\varphi_1(z, a_1),$$

where $\varphi_{10}$ is a constant that does not depend on the distance and the fundamental intensity $\varphi_{10} = \varphi_1(0)$. This approach has been used for the description of two-step cascade processes in Ref. [20].

If we choose $\Delta k_3 \gg \Delta k_2$ the first term of the RHS of Eq. (1b) can be neglected. In zero-order perturbation this equation can be integrated. As a result we have

$$a_2(z) = \frac{2\sigma_5 a_1^2(0)}{\Delta k_2} \sin(\Delta k_2 z/2), \quad (5)$$

$$\cos(\Phi_2) = -\sin(\Delta k_2 z/2). \quad (6)$$

Using the result for $A_2(z)$ Eq. (1c) gives

$$a_3(z) = \frac{2\sigma_3\sigma_5 a_1^3(0)}{(\Delta k_2 + \Delta k_3)\Delta k_2} \sin(\Delta k_2 z/2), \quad (7)$$

$$\cos(\Phi_3) \approx -1. \quad (8)$$

With known $a_2$, $a_3$, $\cos\Phi_2$ and $\cos\Phi_3$ Eq. (3) can be integrated and the result for the nonlinear phase shift $\Delta\varphi_1$ is

$$\Delta\varphi_1 \approx \frac{\sigma_2\sigma_5 a_1^2(0) L}{\Delta k_2} [1 - \mathrm{sinc}(\Delta k_2 L)] + \frac{2\sigma_1\sigma_2\sigma_3\sigma_5 a_1^4(0) L}{\Delta k_2^2(\Delta k_2 + \Delta k_3)} [1 - \mathrm{sinc}(\Delta k_2 L)]. \quad (9)$$

The limitations of application of Eq. (9) come from the restrictions for the wavevector mismatches and the input amplitude. If one takes $\Delta k_3 = 10\,\Delta k_2$ and $\sigma a_1 = \Delta k_2/10$, the result for the NPS will not exceed 0.01 rad. This means that Eq. (9) cannot be used for practical calculations of NPS high enough for all-optical switching purposes, but it is quite useful for physical interpretation of the process of NPS due to multistep cascading.

The first term on the RHS of Eq. (9) (that is the same as obtained in Refs. [19,20] under the condition that $\sigma a_1 \ll \Delta k_2$) is the nonlinear phase shift due to two-step cascading: SHG $(\omega+\omega=2\omega)$ followed by difference frequency mixing $(2\omega-\omega=\omega)$. The second term on the RHS of Eq. (9) accounts for the nonlinear phase shift due to three-step cascading: SHG $(\omega+\omega=2\omega)$, SFG $(2\omega+\omega=3\omega)$, DFG $(3\omega-2\omega=\omega)$. As can be seen the nonlinear phase shift due to three-step cascading is proportional to $|\chi^{(2)}|^4$ and the square of the input intensity. In this way we can consider the three-step contribution (in this low input power limit) as equivalent to the $n_4$ contribution to the nonlinear phase shift in centrosymmetric media. In fact when $\Delta k_2 L = \pm\pi$ (for this values $\Delta\varphi_1$ has extrema) Eq. (9) can be presented as:

$$\Delta\varphi_1 \approx \frac{2\pi}{\lambda}\left[n_2^{(\mathrm{casc})} I - n_4^{(\mathrm{casc})} I^2\right] L, \tag{10}$$

where

$$n_2^{(\mathrm{casc})} = \mathrm{sign}(\Delta k_2)\frac{\lambda\sigma_2^2 L}{c n_1 \varepsilon_0 \pi^2} \tag{11}$$

and

$$n_4^{(\mathrm{casc})} = \frac{12\lambda\sigma_1^2\sigma_2^2 L^2}{(c n_1 \varepsilon_0)^2 \pi^3 \Delta k_3}. \tag{12}$$

Eq. (10) was obtained under the assumption that $\sigma_3 = 3\sigma_1$ and $\sigma_2 = \sigma_5$. From Eq. (10) one can draw the conclusion that the NPS due to three-step cascading does not depend on the sign of the quadratic nonlinearity. The correctness of this conclusion for both three- and four-step cascading was confirmed by the numerical results described below. However, the sign of $n_2^{(\mathrm{casc})}$ and respectively NPS due to second-order cascading can be controlled by the sign of $\Delta k_2$ and $n_4^{(\mathrm{casc})}$ and NPS due to three-step cascading can be controlled by the sign of $\Delta k_3$.

For obtaining the behaviour of the fundamental NPS at arbitrary mismatches $\Delta k_2$ and $\Delta k_3$ and arbitrary input intensity system (1) was solved numerically. We used the following relations between nonlinear coupling coefficients $\sigma_2 = \sigma_5$; $\sigma_3 = 3\sigma_1$; $\sigma_4 = 2\sigma_1$ and $\sigma_2 = \sigma_1$. The results of the exact numerical solution of system (1) for various combinations of the wave vector mismatches $\Delta k_2$ and $\Delta k_3$ are presented in Fig. 2, where the dependence of the nonlinear phase collected by the fundamental wave versus the normalized length of the media for normalized input amplitude of the fundamental wave $\sigma a_{\mathrm{in}} L = 10$ is shown. Dotted line corresponds to the contribution of only two-step cascading and solid line to the full NPS due to coaction of all three channels represented in Fig. 1. As one may see the contribution of the multistep cascading leads to increase of the NPS.

This large nonlinear phase shift makes multistep cascading suitable for low power all-optical switching and signal processing. Fig. 3a, 3b shows the NPS and the depletion of the fundamental wave versus its normalized input amplitude for two different sets of the mismatches

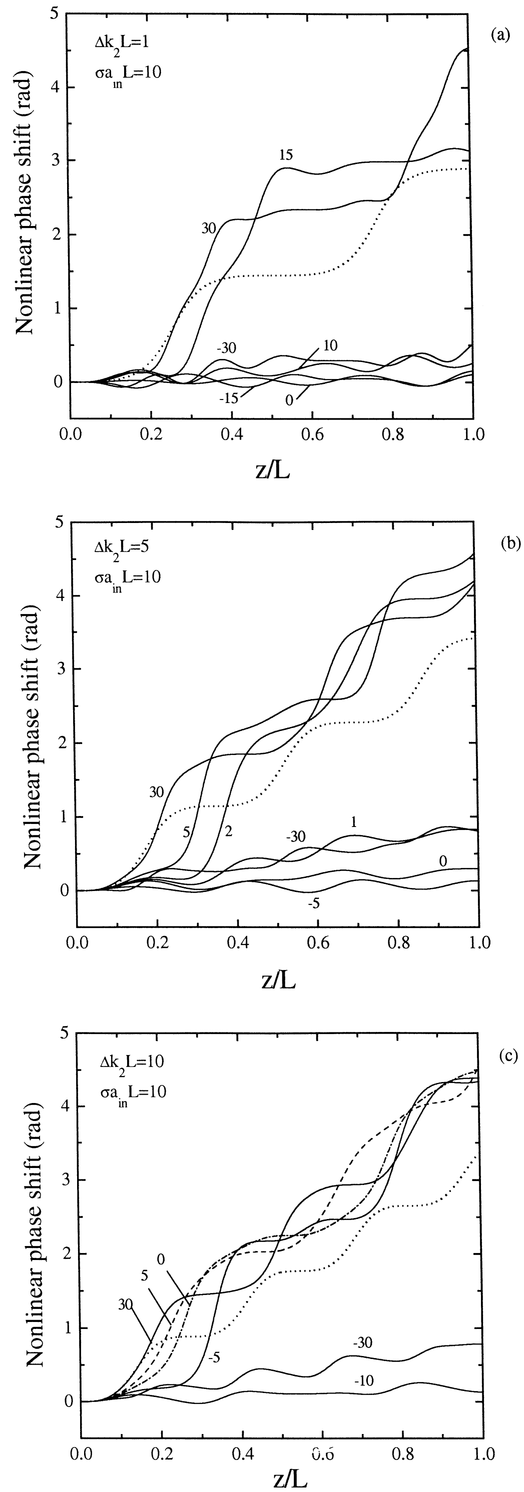


Fig. 2. Nonlinear phase shift of the fundamental wave as a function of the normalized length of the media for (a) $\Delta k_2 L = 1$, (b) $\Delta k_2 L = 5$ and (c) $\Delta k_2 L = 10$. The normalized input amplitude is $\sigma a_1 L = 10$. Dotted line presents the contribution of two step cascading only. The parameter is $\Delta k_3 L$.

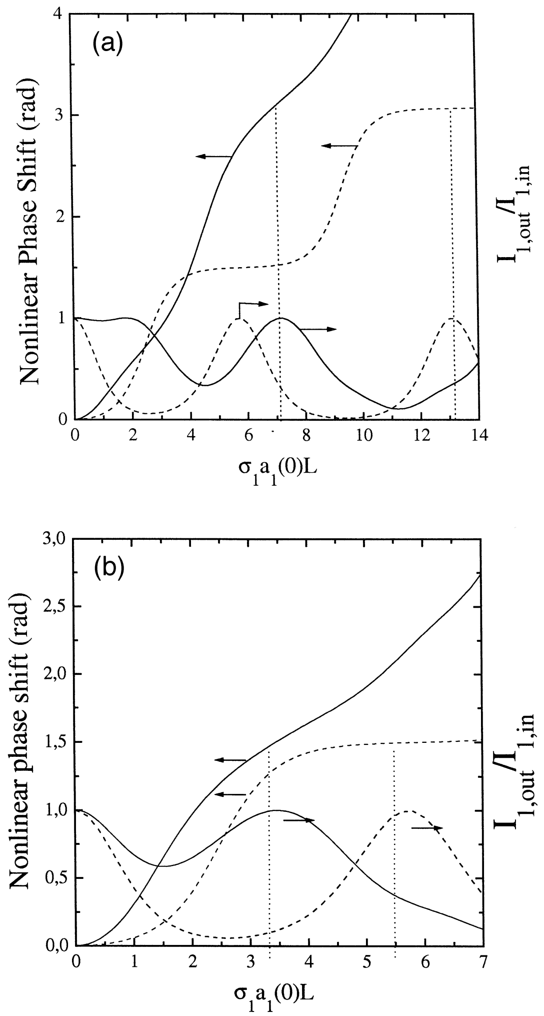


Fig. 3. NPS and the depletion of the fundamental wave as a function of its normalized input amplitude: (a) for the case of multistep cascading with $\Delta k_2 L = 4.8$ and $\Delta k_3 L = 30$ (solid line) and for the case of type I SHG with $\Delta k_2 L = 0.3$ (dashed line); (b) for the case of multistep cascading with $\Delta k_2 L = 2.55$ and $\Delta k_3 L = 15$ (solid line) and for the case of type I SHG with $\Delta k_2 L = 0.3$ (dashed line).

$\Delta k_2$ and $\Delta k_3$, optimal for obtaining an amount of NPS equal to $\pi/2$ or $\pi$ and low losses for the fundamental wave. For comparison in each figure similar optimization for type I SHG ($\Delta k_2 L = 0.3$) is presented. It can be seen that the amplitude required for obtaining these values of the nonlinear phase shift decreases almost two times when using multistep cascading. The value of $\Delta k_2 L$ was found out on the basis of numerical optimization of the two-step process. Our criteria was to find the optimal value of $\Delta k_2 L$ for obtaining NPS equal to 1.5 rad at the middle of the first plateau of 'NPS versus input amplitude' dependence. The value of $\Delta k_2 L = 0.3$ used here is different from the optimal value cited in Refs. [1,2] because the analysis done in these works is valid only for low input amplitudes ($\sigma a_1/\Delta k_2 \ll 1$).

The dependences shown in Figs. 2 and 3 are for relatively small values of the normalized mismatches $\Delta k_2 L$ and $\Delta k_3 L$. When $\Delta k_2 \gg \Delta k_3$ or $\Delta k_3 \gg \Delta k_2$ the resulting NPS is close to the NPS due to the process of SHG only. This means that substantial increase of the NPS due to multistep $\chi^{(2)}$ cascading is possible when both SHG and SFG processes are nearly phase matched.

In conclusion, here we present a method for obtaining a large NPS for a fundamental wave that passes through quadratic nonlinear media. The presence of multistep $\chi^{(2)}$ cascading leads to increase of the value of the NPS in comparison with two-step cascading. This more efficient way of generating a large nonlinear phase shift can be used for reduction of the switching intensity of existing all optical switching devices based on $\chi^{(2)}$ cascading [3,6] and for construction of intracavity nonlinear optical devices for mode locking [21–23].

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