High-order nonlinear phase shift caused by cascaded third-order processes

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We show, for the first time to our knowledge, that the fundamental beam that participates in the process of third-harmonic generation experiences an additional high-order nonlinear phase shift. The magnitude of the shift is proportional to the square of the pump intensity and the length of the sample and depends on the deviation from the exact phase-matched condition. This phase shift arises from cascaded third-order processes. Its value can exceed the value of the phase shift that originates from inherent fifth-order susceptibility of the nonlinear medium. Its sign is controllable by the sign of the phase mismatch of the third-harmonic generation process.

In general, the phase shift that a wave propagating through a nonlinear medium suffers is described by the intensity-dependent refractive index[1]:

$$n = n_0 + n_2 I + n_4 I^2 + n_6 I^3 + \dots . \quad (1)$$

The first, second, and third nonlinear terms on the right-hand side of Eq. (1) are connected to the cubic-, fifth-, and seventh-order nonlinear processes, respectively:

$$n_2 = 3\chi^{(3)}/(4\epsilon_0 c n_0^2), \qquad n_4 = 5\chi^{(5)}/(4\epsilon_0^2 c^2 n_0^3),$$
$$n_6 = 35\chi^{(7)}/(16\epsilon_0^3 c^3 n_0^4). \quad (2)$$

Recently it was shown[2] that the n_2 coefficient in quadratic media is in fact a sum of two terms, one that is due to the inherent $\chi^{(3)}$ susceptibility of the media and the second to the cascading of the second-order nonlinearity: $n_2 = n_2^{\text{dir}} + n_2^{\text{casc}}$. The cascaded term is comparable with or higher in value than n_2^{dir} only when the beam is involved in a nearly phase-matched second-order nonlinear-optical process.

One should expect the cascading of third-order nonlinearities to influence the nonlinear phase modulation of the fundamental beam in the same way as the cascaded second-order processes do. Until now this problem was investigated only with respect to the efficiency of the generated waves.[3,4] To our knowledge an analysis of self- and cross-phase modulation of the fundamental beam owing to cascading of third-order nonlinearities has not been made.

Here we present a study of the nonlinear self-phase modulation of a fundamental beam involved in a slightly mismatched process of third-harmonic generation (THG). We show that in this case n_4 consists of two terms: n_4^{dir} proportional to the corresponding component $\chi^{(5)}$ of the fifth-order nonlinear susceptibility tensor and a cascaded term n_4^{casc} proportional to $(\chi^{(3)})^2$. By appropriate choice of the sample length, wave-vector mismatch, and beam intensity the value of n_4^{casc} can always be made to exceed that of n_4^{dir}.

We start our investigation of a process of THG ($\omega_3 = \omega_1 + \omega_1 + \omega_1$) in a nonlinear crystal of length L. It is assumed that the fundamental and the generated waves are linearly polarized and that the electric field is

$$\mathbf{E} = 1/2\{\hat{e}_1 A_1 \exp[-i(\omega t - k_1 z)]$$
$$+ \hat{e}_3 A_3 \exp[-i(3\omega t - k_3 z)] + \text{c.c.}\}, \quad (3)$$

where $k_j = 2\pi n_j/\lambda_j$ and ω_j are the corresponding propagation constants and frequencies ($\hat{e}_1$ and $\hat{e}_2$ are the polarization unit vectors). $A_j\,(j = 1, 3)$ are the complex slowly varying wave amplitudes. They incorporate both the real amplitudes a_j and the phases φ_j of the waves: $A_j(z) = a_j(z)\exp[i\varphi_j(z)]$. The two waves are considered to be nearly phase matched, which can be achieved by use of natural birefringence or the quasi-phase-matched (QPM) technique.[5] The QPM technique was developed for second-order nonlinear processes, but it has recently been extended, both theoretically and experimentally, to the case of THG.[6]

Starting from Maxwell's equations and after accounting for all relevant cubic and fifth-order terms in the nonlinear polarization, we drive the slowly varying envelope equations that describe the THG in a transparent nonlinear medium in the form

$$\frac{dA_1}{dz} = i(\gamma_1|A_1|^2 + \gamma_2|A_3|^2 + \delta|A_1|^4)$$
$$\times A_1 + i\gamma_{13}A_1^{*2}A_3 \exp(i\Delta kz),$$
$$\frac{dA_3}{dz} = i\gamma_4|A_1|^2 A_3 + i\gamma_{31}A_1^3 \exp(-i\Delta kz), \quad (4)$$

where the nonlinear coupling coefficients are specified in Table 1. In the case of QPM technique the

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Table 1. Nonlinear Coupling Coefficients

$$\gamma_1 = \frac{\omega}{c}\,\frac{3}{8n_0(\omega)}\,\hat{e}_1 \cdot \chi^{(3)}\vdots \hat{e}_1\hat{e}_1\hat{e}_1,$$

$$\gamma_2 = 2\,\frac{\omega}{c}\,\frac{3}{8n_0(\omega)}\,\hat{e}_1 \cdot \chi^{(3)}\vdots \hat{e}_3\hat{e}_3\hat{e}_1,$$

$$\delta = \frac{\omega}{c}\,\frac{5}{16n_0(\omega)}\,\hat{e}_1 \cdot \chi^{(5)}\vdots \hat{e}_1\hat{e}_1\hat{e}_1\hat{e}_1\hat{e}_1,$$

$$\gamma_{13} = \frac{\omega}{c}\,\frac{3}{8n_0(\omega)}\,\hat{e}_1 \cdot \chi^{(3)}\vdots \hat{e}_1\hat{e}_1\hat{e}_3$$

$$\gamma_{31} = \frac{3\omega}{c}\,\frac{3}{8n_0(3\omega)}\,\hat{e}_3 \cdot \chi^{(3)}\mathrm{L}\hat{e}_1\hat{e}_1\hat{e}_1$$

$$\gamma_4 = 2\,\frac{3\omega}{c}\,\frac{3}{8n_0(3\omega)}\,\hat{e}_3 \cdot \chi^{(3)}\vdots \hat{e}_1\hat{e}_1\hat{e}_3$$

wave-vector mismatch is $\Delta k = k_3 - 3k_1 - K$, where $K = 2\pi/T$ and T is the periodicity of the grating.[5,6]

The system of Eqs. (4) was solved analytically by use of the fixed intensity approximation. This approximation, initially developed for the description of type I second-harmonic generation[7] and applied later for type II second-harmonic generation,[8,9] suggests no depletion for the intensity of the fundamental waves but a possible change of their phases. Here the fixed intensity approximation approach is extended to the case of THG. Following this method we reduce the system of Eqs. (4) to an ordinary differential equation for the complex amplitude of the third harmonic wave:

$$\frac{\mathrm{d}^2 A_3}{\mathrm{d}z^2} + i\Delta\,\frac{\mathrm{d}A_3}{\mathrm{d}z} + SA_3 = 0\,, \tag{5}$$

in which (after assuming that $|A_3|^2 \ll |A_1|^2 =$ constant) the coefficients Δ and S are specified as $\Delta = \Delta k - (\gamma_4 + 3\gamma_1)|A_1|^2$ and $S = [\Delta k\gamma_4 + 3(\gamma_{13}\gamma_{31} - \gamma_1\gamma_4)|A_1|^2]|A_1|^2$. The integration of Eq. (5) gives

$$a_3(z) = \gamma_{31}{a_1}^3\ \mathrm{sinc}(\Lambda z)z,$$

$$\varphi_3(z) = (\pi/2) + 3\varphi_{10} - (\Delta/2)z\,, \tag{6}$$

where $\Lambda^2 = \Delta^2/4 + S$ and φ_{10} is the initial phase of the fundamental wave. As $\mathrm{d}\varphi_3/\mathrm{d}z = -(\Delta/2)$, and on the basis of Eqs. (4), we obtain the following equation for the phase of the fundamental beam:

$$\frac{\mathrm{d}\varphi_1}{\mathrm{d}z} = \gamma_1{a_1}^2 + \delta{a_1}^4 + \gamma_2{a_3}^2 + (\gamma_{13}/\gamma_{31})(a_3/a_1)^2(\Delta/2 + \gamma_4{a_1}^2)\,. \tag{7}$$

As a result the nonlinear phase shift (NLPS) $\Delta\varphi_1 = \varphi_1(L) - \varphi_{10}$ of the fundamental wave at the output $(z = L)$ of the sample is obtained in the form

$$\Delta\varphi_1 = (\gamma_1{a_1}^2 + \delta{a_1}^4)L - [\,\gamma_{13}\gamma_{31}(\Delta/2) - \gamma_{31}(\gamma_2\gamma_{31} - \gamma_4\gamma_{13}){a_1}^2]\frac{{a_1}^4 L}{2\Lambda^2}[1 - \mathrm{sinc}(2\Lambda L)]\,. \tag{8}$$

The first term in Eq. (8) represents the contribution of the direct third- and fifth-order processes to the NLPS. The second one, which we denote $\Delta{\varphi_1}^{\mathrm{casc}}$, accounts for the self-phase modulation of the beam that is due to the cascaded third-order processes. The physical explanation of the cascaded third-order NLPS is based on the interference of the fundamental wave and the wave generated as a result of the four-wave mixing process: $\omega = 3\omega - \omega - \omega$.

Figure 1 illustrates the dependence of the cascaded part $\Delta{\varphi_1}^{\mathrm{casc}}$ of the NLPS [Eq. (8)] of the fundamental beam as a function of the normalized phase mismatch ΔkL for different values of the normalized input intensity $\gamma_{13}|A_1|^2L$. Assuming that the medium is transparent for both the fundamental and the generated wave frequencies, we accept here (see Table 1) the following equalities[10]: $\gamma_{31} \approx \gamma_{13}$ and $\gamma_4 \approx 3\gamma_2$. In our calculations we took that $\gamma_1 = \gamma_{13}$ and $\gamma_4 = 6\gamma_{13}$. The phase-shift curves are centered at $\Delta kL = 0$ only for low values of the input intensity. For higher values of $\gamma_{13}|A_1|^2L$ (unlike in the corresponding case of second-harmonic generation) the curves are no longer centered at the point $\Delta kL = 0$, and the maximum and the minimum absolute values differ. This fact is due to the correct accounting for all relevant nonlinear terms in our model and can be quite important in experimental situations when a maximum NLPS is needed.

On the basis of Table 1 and using Eqs. (2) and (8), we define the cascaded part of nonlinear refractive

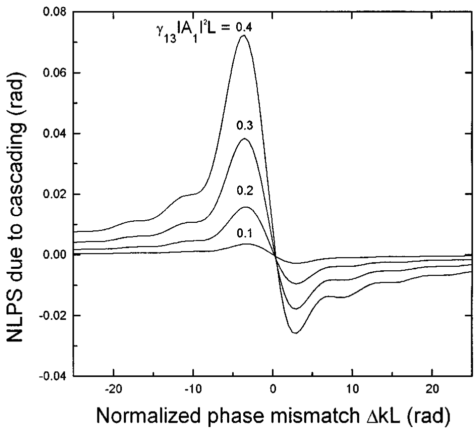

Fig. 1. High-order NLPS (owing to cascading) as a function of the normalized phase mismatch ΔkL. The parameter is the value of the normalized input intensity $\gamma_{13}|A_1|^2L$.

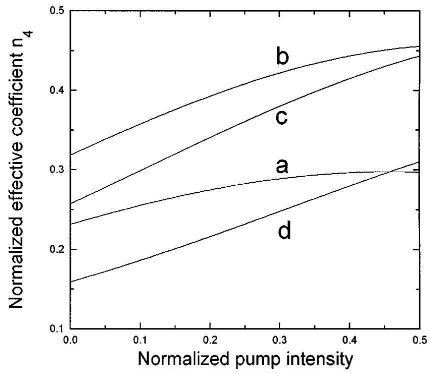

Fig. 2. Normalized effective coefficient $\tilde{n}_4$ as a function of the normalized pump intensity $\gamma_{13}|A_1|^2L$. The parameter is the normalized phase mismatch ΔkL (in radians) with the following values: a, $-\pi/2$; b, $-\pi$; c, $-3\pi/2$; d, -2π.

Table 2. Numerical Values of the Nonlinear Refractive-Index Coefficients n_2, n_4, and $n_{4,\max}^{\mathrm{case}}$ for Various Materials and Wavelengths[a]

Material	λ (nm)	n_2 (cm^2/W)	n_4 (cm^4/W^2)	$n_{4,\max}^{\mathrm{case}}$ (cm^4/W^2)	Linear Loss, TPA	Reference
RG 780	740	5.6×10^{-13}	-8.4×10^{-22}	-8.5×10^{-21}	Negligible	11
PTS	1064	5.0×10^{-12}	-5.0×10^{-21}	-4.7×10^{-19}	Significant	12
PTS	1600	2.2×10^{-12}	-8.0×10^{-22}	-6.0×10^{-20}	Negligible	13
AlGaAs	1550	1.5×10^{-13}	-5.0×10^{-23}	-2.9×10^{-22}	Negligible	14

[a] $n_{4,\max}^{\mathrm{case}}$ is calculated with Eq. (10) for $L = 1$ cm and $\Delta kL = \pi$; TPA is two-photon absorption.

coefficient n_4 in the form

$$n_4{}^{\mathrm{casc}} = \frac{\omega}{c} n_2{}^2 \frac{\Delta}{4\Lambda^2}[1 - \mathrm{sinc}(2\Lambda L)]. \tag{9}$$

The value of $n_4{}^{\mathrm{casc}}$ increases with increase of the input fundamental wave intensity. This fact is illustrated on Fig. 2, where the normalized fifth-order cascaded nonlinear refractive-index coefficient $\tilde{n}_4 = (n_4{}^{\mathrm{casc}}/n_2{}^2)/(\lambda/2\pi L)$ is plotted against the normalized pump intensity $\gamma_{13}|A_1|^2 L$ for several values of the normalized phase mismatch ΔkL. The variation range of the normalized pump intensity is chosen in such a way that the depletion of the fundamental beam does not exceed 10%. For higher values of the intensity the fixed pump intensity approximation is no longer valid, and one should use a numerical solution of Eqs. (4). Nevertheless, the approach that we present here seems to be a good analytical tool, providing an instructive and comprehensive physical understanding of the problem.

For $\gamma_{13}|A_1|^2 L \ll 1$, Eq. (9) has extrema at $2\Lambda L \approx \Delta L \approx \Delta kL = \pm\pi$ that have the form

$$n_4{}^{\mathrm{casc}}(2\Lambda L = \pm\pi) = -\mathrm{sgn}(\Delta k) 2 n_2{}^2 L/\lambda. \tag{10}$$

It is seen that by choosing the sign of the mismatch Δk we can choose the sign of $n_4{}^{\mathrm{casc}}$ and also that we can effectively increase its value by increasing the length of the nonlinear medium. To obtain some numbers we took the available data for n_2 and n_4 for the materials[11–14] listed in Table 2. For example, for polydiacetylene *para*-toluene sulfonate (PTS) at $\lambda = 1600$ nm,[13] assuming that $L = 1$ cm, we get $n_{4,\max}^{\mathrm{casc}} = \pm 6 \times 10^{-20}$ cm^4/W^2, which is almost a 2-order-higher value than the reported direct one. This numerical example shows a more realistic situation for experimental observation of bistable solitary waves, for which the ratio between the third- and the fifth-order coefficients is an essential parameter.[15–17] The relatively high values of this parameter provide a larger range of bistability of the solitary waves. PTS (for which the quintic nonlinearity contribution is significant) is also believed to present a unique opportunity for studies of stable two-dimensional solitary waves.[13,18] As shown in Ref. 13, the lower the ratio of n_2/n_4, the lower the required power for stable beam propagation.

In conclusion, we have studied analytically the high-order NLPS of the fundamental wave involved in the process of type I THG in media with centers of inversion. The nonlinear medium should permit phase matching of the process by use of natural birefrigence or the so-called QPM techniques. The high-order NLPS that is due to the cascaded third-order nonlinearities is controllable in sign by proper choice of the sign of the phase mismatch between the fundamental and the third-harmonic waves. A similar analysis can be performed for studying direct THG in quadratic media. In this case three- and four-step cascaded processes will also contribute to the magnitude of $n_4{}^{\mathrm{casc}}$.

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