Nonlinear phase shift as a result of cascaded third-order processes

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In centrosymmetric media, in addition to the third- and fifth-order nonlinearity, a cascaded $\chi^{(3)}:\chi^{(3)}$ nonlinearity contributes to the nonlinear refraction. The described effect takes place when the input beam is involved in nearly phase-matched third-harmonic generation. The nonlinear phase shift produced by this new cascaded nonlinearity is comparable to the nonlinear phase shift caused by the direct third-order nonlinearity $\chi^{(3)}$. A new analytical approach, low depletion approximation, was developed and used to study the cascaded third-order processes in the conditions of not very high third-harmonic conversion coefficients. At higher conversion coefficients the effect was studied by numerical simulations. The presence of a seeded third-harmonic wave gives another possibility of controlling the additional phase shift of the fundamental wave. The conditions for stationary copropagation of the fundamental and the generated wave are presented. [S1050-2947(98)01004-X]

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I. INTRODUCTION

An intense wave propagating through nonlinear media collects a nonlinear phase shift (NPS) that is described by the expansion of the refractive index on powers of the light intensity I:

$$n=n_0+n_2I+n_4I^2+n_6I^3+\cdots . \tag{1}$$

The $\chi^{(3)}$ nonlinear refractive index n_2 $[n_2=3\chi^{(3)}/(4\varepsilon_0cn_0^2)]$ has a key role for such nonlinear optical processes as optical bistability [1], all optical switching [2], self-focusing, and self-defocusing [3]. Recently it was shown that in noncentrosymmetric media it is possible to control the magnitude and the sign of the effective n_2 by exploring the so-called cascaded second-order processes [4–9]. The value of this ''cascade type'' $n_{2,\mathrm{casc}}$ is as a rule higher than the nonlinear index of refraction that is connected with the inherent cubic nonlinearity of the media. The $n_{2,\mathrm{casc}}$ presentation of the additional phase shift due to $\chi^{(2)}:\chi^{(2)}$ cascaded processes is valid only for relatively low input intensities; at higher intensities, however, the NPS becomes asymptotically linear in the field [4,7]. Is this change of the slope of the ''NPS versus irradiance'' dependence a generic behavior for all cascade nonlinearities? We will try to answer this question by exploring the next order cascade nonlinearity.

The second nonlinear constant in Eq. (1), n_4, known as the $\chi^{(5)}$ nonlinear index of refraction $[n_4=5\chi^{(5)}/(4\varepsilon_0^2c^2n_0^3)]$, is also an important parameter in contemporary nonlinear optics. Theoretical investigations show that in order to obtain spatial solitary waves a certain relation between n_2 and n_4 must imply [10,11]. In particular, in [10] it is shown that the lower the ratio of n_2/n_4, the lower is the power for stable beam propagation. In analogy with the opportunity to control the magnitude and the sign of the n_2 in noncentrosymmetric media one can ask: is it possible to control the magnitude and the sign of n_2 and n_4 in centrosymmetric media? As we have found this can be done using cascaded third-order nonlinear processes.

The influence of cascaded third-order processes (CTOP) on the amplitude of the generated wave by six photon phase matched nonlinear interactions was analyzed for the first time 20 years ago in [12]. This role of CTOP was described in detail in Refs. [3,13,14]. The main conclusion resulting from these investigations is that the efficiency of the six-photon phase matched processes is proportional to the square of the effective fifth-order nonlinearity, which is a sum of two terms:

$$\chi_{\mathrm{eff}}^{(5)}=\chi_{\mathrm{int}}^{(5)}+\chi_{\mathrm{casc}}^{(5)}. \tag{2}$$

The second term is a result of a consequence of two third-order processes $[\chi_{\mathrm{casc}}^{(5)}=\mathrm{const}\times\chi^{(3)}\cdot\chi^{(3)}]$. From Eq. (2), by measuring $\chi_{\mathrm{eff}}^{(5)}$ and by calculating $\chi_{\mathrm{casc}}^{(5)}$ it is possible to determine $\chi_{\mathrm{int}}^{(5)}$ [12,15]. At that time no attention was paid to the influence of the CTOP on the phases of the fundamental waves. In Ref. [16] we published our preliminary investigation of the phase modulation of the fundamental wave involved in the process of low efficiency third-harmonic generation. Shortly after that the effect of CTOP on the structure and stability of spatial solitary waves was studied in [17].

In this paper we consider the process of nearly phase-matched type-I efficient third-harmonic generation in centrosymmetric media and in more detail the effect of NPS of the intense fundamental wave that arises as a result of CTOP. At these conditions, the consideration of the effect of self-phase modulation of the third-harmonic wave is important and is taken into account. The presence of seeding (nonzero input at third-harmonic frequency) is also considered. We show that the magnitude and the phase of the seeded wave gives an additional degree of freedom for controlling the NPS of the fundamental wave. The conditions for stationary copropagation of the fundamental and the third-harmonic wave are defined following the approach developed for the case of second-harmonic generation [18–20].

II. PLANE-WAVE EQUATIONS

The amplitude equations that describe the process of type-I third-harmonic generation $(\omega_3=\omega_1+\omega_1+\omega_1)$ in lossless cubic media for linear polarized plane waves are [21]:

$$\frac{dA_1}{dz}=i(\gamma_1|A_1|^2+\gamma_2|A_3|^2)A_1+i\gamma_3A_1^{*2}A_3\exp(i\Delta kz),$$

$$\frac{dA_3}{dz}=i(\gamma_4|A_1|^2+\gamma_5|A_3|^2)A_3+i\gamma_3A_1^3\exp(-i\Delta kz). \quad (3)$$

The phase velocity mismatch is $\Delta k=k_3-3k_1$. In case of using the quasi-phase-matched technique [22,23] the wave vector mismatch is $\Delta k=k_3-3k_1-K$, where $K=2\pi/T$ and T is the periodicity of the grating. γ_j are the nonlinear coupling coefficients that include convolutions of the third-order susceptibility tensor $\chi^{(3)}$ with the polarization vectors of the interacting waves [16,24]. A_j are the complex amplitudes that incorporate both the real amplitudes $|A_j|$ and the phases φ_j of the waves: $A_j=|A_j|\exp(i\varphi_j)$.

It is convenient to work with normalized waves amplitudes defined by $f=|A_1|/u$ and $h=|A_3|/u$, where u is determined by the boundary conditions $u=\sqrt{|A_1(0)|^2+|A_3(0)|^2}$. Then Eqs. (3) can be rewritten as

$$-\frac{df^2}{d\xi}=\frac{dh^2}{d\xi}=2pf^3h\ \sin\Phi, \quad (4a)$$

$$\frac{f}{p}\frac{d\varphi_1}{d\xi}=\tilde{\gamma}_1f^3+\tilde{\gamma}_2fh^2+f^2h\ \cos\Phi, \quad (4b)$$

$$\frac{h}{p}\frac{d\varphi_3}{d\xi}=\tilde{\gamma}_4f^2h+\tilde{\gamma}_5h^3+f^3\ \cos\Phi. \quad (4c)$$

In Eq. (4) $p=\gamma_3u^2L$ is the normalized input intensity, $\tilde{\gamma}_j=\gamma_j/\gamma_3$ $(j=1,2,4,5)$, $\Phi=\varphi_3-3\varphi_1+\Delta kL\xi$, and $\xi=z/L$ is the normalized distance with L—the length of the nonlinear media.

The system (4) has two invariants:

$$f^2+h^2=1$$

and

$$f^3h\ \cos\Phi+\Lambda h^2+\tfrac{1}{4}\Delta\gamma h^4=\Gamma_0. \quad (5)$$

The following notations have been introduced in Eq. (5): the normalized mismatch $\Delta\tilde{k}=\Delta k/\gamma_3u^2$, $\Lambda=0.5(\tilde{\gamma}_4-3\tilde{\gamma}_1+\Delta\tilde{k})$, and $\Delta\gamma=\tilde{\gamma}_5-\tilde{\gamma}_4+3(\tilde{\gamma}_1-\tilde{\gamma}_2)$.

There are three approaches to solving Eq. (4). The first is to obtain formula for f, h, φ_1, and φ_3 expressed in special functions without any approximations as is done for cascaded second-order processes [19,25]. The second approach is to numerically solve the system and the last one is to use an approximation that is valid only for low levels of the efficiency of the third-harmonic generation process. We developed here low depletion approximation, described in the next section. The numerical approach was applied for the case of high-efficiency third-harmonic generation.

III. LOW DEPLETION APPROXIMATION

A. Nonlinear phase shift due to CTOP

The use of the approximation valid for low depletion of the fundamental wave can be justified by two arguments. First, it allows the obtaining of simple analytical formulas suitable for analyzing the physical background of the problem. Second, this approximation is valid only for the description of nonlinear optical processes with low conversion coefficients. This is exactly the case of third-harmonic generation in most of the nonlinear media. The reported third-harmonic generation efficiency coefficients do not exceed several percent even for the highest possible nondamaging intensities [3,15,26–30].

The low depletion (LD) approximation described here is an extension of the fixed intensity approximation developed for description of second harmonic generation [31–33]. The fixed intensity approximation suggests no depletion of the intensity of the fundamental wave $[f(\xi)=f_0]$, but a possible change of its phase. In the LD approximation presented here we suggest not only possible changes of the phases of the interacting waves, but also the possibility of not very strong depletion of the fundamental wave $(f^2\geqslant 0.8,\ h^2\leqslant 0.2)$.

Using Eq. (5) and neglecting the terms that are proportional to h^6 and h^8, an expression for $f^3h\ \sin\Phi$ can be found:

$$f^3h\ \sin\Phi=\sqrt{-Q^2h^4+Vh^2-\Gamma_0}, \quad (6)$$

where $Q^2=3+\Lambda^2-0.5\Gamma_0\Delta\gamma$ and $V=1+2\Gamma_0\Lambda$.

Then integrating Eq. (4a) we obtain for the intensity of the third-harmonic signal:

$$h^2(\xi)=\frac{1}{2Q^2}[V-N\ \cos(2pQ\xi)-S\ \sin(2pQ\xi)]. \quad (7)$$

In Eq. (7) $S=2Qf_0^3h_0\sin(\Phi_0)$, $N=V-2Q^2h_0^2$, $\Phi_0=\varphi_3(0)-3\varphi_1(0)$, and h_0 and f_0 are the input normalized amplitudes for the third-harmonic and fundamental waves.

In case of no seeding, $h_0=0$, and

$$h^2(z)=\gamma_3^2u^4z^2\text{sinc}^2\left(\frac{\pi}{2}\frac{z}{l_{\text{coh}}}\right). \quad (8)$$

In Eq. (8) sinc$x$ replaces sin$x$/x and the coherence length l_{coh} of the third-harmonic generation process was defined, $l_{\text{coh}}=\pi/2\gamma_3u^2Q$.

Using again Eq. (5) and keeping only terms proportional to h^n $(n\leqslant 4)$ we obtain from Eq. (4b)

$$\frac{1}{p}\frac{d\varphi_1}{d\xi}=\tilde{\gamma}_1+\Gamma_0+\alpha_1 h^2+\alpha_2 h^4. \quad (9)$$

Here $\alpha_1=[\Gamma_0-\Lambda+(\tilde{\gamma}_2-\tilde{\gamma}_1)]$ and $\alpha_2=[\Gamma_0-\Lambda-(\Delta\gamma/4)]$.

Integration of Eq. (9) with the use of Eq. (7) gives, for the nonlinear phase shift

$$\Delta\varphi_1=\varphi_1(\xi)-\varphi_1(0),$$

$$\Delta\varphi_1=p(\tilde{\gamma}_1+\Gamma_0)\xi+\frac{\alpha_1 p\xi}{2Q^2}\{V-N\ \mathrm{sinc}(2pQ\xi)-SpQ\xi\ \mathrm{sinc}^2(pQ\xi)\}+\frac{\alpha_2 p\xi}{4Q^4}\left\{V^2+\frac{N^2+S^2}{2}+\frac{N^2-S^2}{2}\mathrm{sinc}(4pQ\xi)-2VN\ \mathrm{sinc}(2pQ\xi)+2pSQ\xi[N\ \mathrm{sinc}^2(2pQ\xi)-V\ \mathrm{sinc}^2(pQ\xi)]\right\}. \quad (10)$$

For the case of no third-harmonic signal at the input of the nonlinear media,

$$\Delta\varphi_1=p\tilde{\gamma}_1\xi+\frac{\alpha_1 p\xi}{2Q^2}[1-\mathrm{sinc}(2pQ\xi)]+\frac{\alpha_2 p\xi}{8Q^4}[3+\mathrm{sinc}(4pQ\xi)-4\ \mathrm{sinc}(2pQ\xi)]. \quad (11)$$

The range of validity of the derived formulas was verified by comparison with the exact numerical calculations, as shown in the following sections. As we already mentioned, the depletion of the fundamental wave (respectively, the conversion coefficient into third harmonic) should not exceed 20%.

B. Physical explanation of the phase modulation by CTOP: Definition of $\chi^{(5)}_{\mathrm{casc}}$

For very low input intensities $p\ll|\Delta k|L$ the normalized mismatch is $|\Delta\tilde{k}|\gg 1$ and we can set $\alpha_1/2Q^2\approx -1/2\Lambda\approx -\gamma_3 u^2/\Delta k$. Then expression (11) is simplified to

$$\Delta\varphi_1=\gamma_1 u^2 z-\frac{\gamma_3^2}{\Delta k}u^4 z[1-\mathrm{sinc}(\Delta kz)]. \quad (12)$$

The first term in Eq. (12) is a result of direct one-step processes of self-phase-modulation described by the natural $\chi^{(3)}$ susceptibility of the media. The second term in Eq. (12) is a result of two-step CTOP ($\chi^{(3)}:\chi^{(3)}$ cascading). The physical explanation of the process of the NPS due to the CTOP is based on the interference of three waves (see Fig. 1): (i) the linearly shifted fundamental wave, (ii) the wave generated as a result of a single-step third-order processes—degenerate and nondegenerate four-wave-mixing processes, and (iii) the wave generated as a result of the two-step nondegenerate four-wave-mixing interactions $\omega=3\omega-\omega-\omega$. The phase shift for the waves (ii) and (iii) is different from that of linearly shifted wave (i). The amplitude of waves (ii) and (iii) depends nonlinearly on the input wave amplitude. As a result the output wave has intensity dependent NPS. The schematic drawing shown in Fig. 1 refers to third-harmonic generation with zero input third-harmonic signal ($h_0=0$). In fact, the phase shift due to cascaded third-order processes (the lower channel in Fig. 1) is a result of effective fifth-order process with $\chi^{(5)}_{\mathrm{casc}}\propto(\chi^{(3)})^2$.

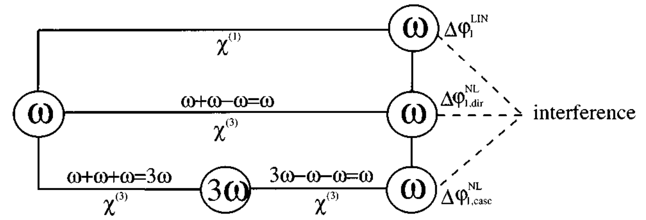

FIG. 1. Schematic drawing for illustration of the physical explanation of the process of formation of a nonlinear phase shift in nearly phase-matched third-harmonic media in the case of a zero input third-harmonic signal. The upper channel represents the linear phase shift of the input wave, the middle channel the generation of the wave by degenerate four-wave mixing, and the lower channel the generation of the wave by two-step third-order processes.

Taking into account that $\gamma_3=(3\omega/c)(1/8n_0)\chi^{(3)}$ we obtain that the NPS due to $\chi^{(3)}:\chi^{(3)}$ cascading is described by the effective fifth-order nonlinearity,

$$\chi^{(5)}_{\mathrm{casc}}=-\frac{9\pi}{10}|\chi^{(3)}|^2\frac{1}{\Delta kL}\frac{L}{n\lambda}. \quad (13)$$

The contribution of $\chi^{(5)}_{\mathrm{eff}}$ can exceed the contribution of the intrinsic $\chi^{(5)}_{\mathrm{int}}$ of the media. For example, for the Shott glass RG 780, for which $\chi^{(5)}_{\mathrm{int}}$ and $\chi^{(3)}_{\mathrm{int}}$ are known [34], we estimated that $\chi^{(5)}_{\mathrm{casc}}/\chi^{(5)}_{\mathrm{int}}=11.4$ (for this estimation $\Delta kL=\pi$ and $L/n\lambda=1000$ were taken).

For this range of input intensities the cascaded NPS can be presented as depending on an effective $n_{4,\mathrm{casc}}$ that has the form

$$n_{4,\mathrm{casc}}=-\frac{2n_2^2}{\Delta k\lambda}. \quad (14)$$

It is interesting to point out that the cascade-type fifth-order nonlinearity responsible for the fifth-harmonic generation has the same structure as expression (13) [3,12–14]. However, $\chi^{(5)}_{\mathrm{casc}}$ responsible for obtaining NPS is much higher in value in comparison with $\chi^{(5)}_{\mathrm{casc}}$, responsible, for example, for the fifth-harmonic generation because of the fact that in the last case the mismatch Δk is some orders of magnitude larger.

Let us now consider the opposite case for the input fundamental intensity, $p\geq|\Delta k|L$, then the normalized mismatch $\Delta\tilde{k}$ can be neglected in the expression for Λ. For this high input intensity limit Λ, Q, α_1, and α_2 are constants that depend only on the nonlinear coupling coefficients γ_i. Then the cascaded part of the NPS, $\Delta\varphi_{1,\mathrm{casc}}=\Delta\varphi_1-\gamma_1 u^2 z$, is

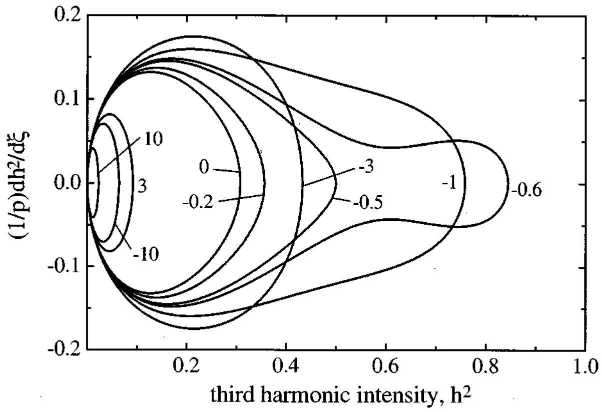

FIG. 2. Phase-plane portraits for nonseeded third-harmonic generation for various normalized mismatches $\Delta\widetilde{k}$.

$$\Delta\varphi_{1,\mathrm{casc}}=\frac{1}{2Q^2}\left(\alpha_1+3\,\frac{\alpha_2}{2Q^2}\right)\gamma_3 u^2 z$$
$$-\frac{1}{2Q^3}\left\{\alpha_1+\frac{\alpha_2}{Q^2}\left[1+\frac{1}{8}\cos\!\left(\pi\,\frac{z}{l_{\mathrm{coh}}}\right)\right]\right\}$$
$$\times\sin\!\left(\pi\,\frac{z}{l_{\mathrm{coh}}}\right). \qquad (15)$$

From Eq. (15) it can be seen that for this intensity range the cascaded NPS depends linearly on the input intensity. The last term in Eq. (15) causes small steps. The cascaded NPS, $\Delta\varphi_{1,\mathrm{casc}}$, has the same behavior as the NPS caused by the natural $\chi^{(3)}$ of the media. The numerical calculations described in the next section confirmed that for higher values of p the cascaded NPS has a tendency to saturate and is proportional to the square of the input fundamental amplitude. In this case the additional phase shift should be described by an effective $n_{2,\mathrm{casc}}$.

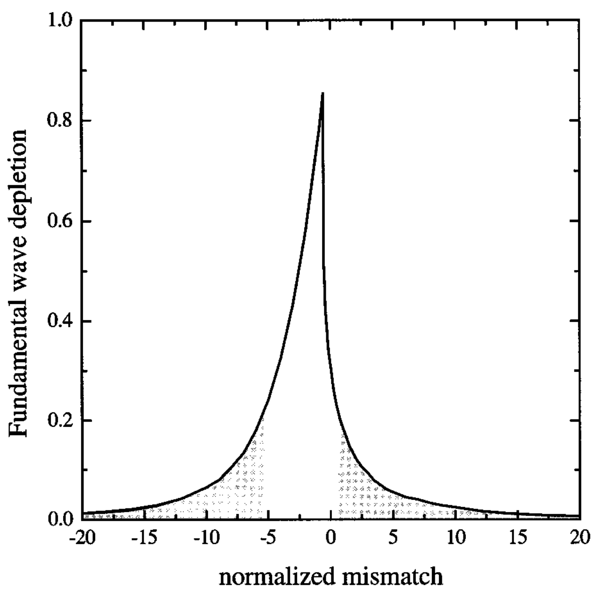

FIG. 3. Periodic depletion of the fundamental wave intensity (respectively, the pick to pick change of the third-harmonic intensity) as a function of the normalized mismatch $\Delta\widetilde{k}=\Delta k/\gamma_3 u^2$, as found from Eq. (16). The patterned sectors give the initial conditions for which the low depletion approximation should be used.

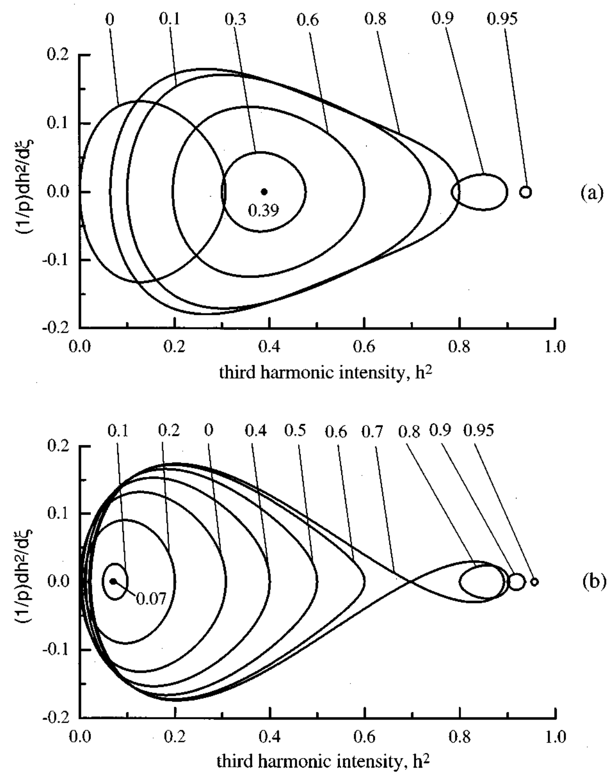

FIG. 4. Phase-plane portraits for seeded third-harmonic generation with $\Phi_0=0$ (a), $\Phi_0=\pi$ (b), and $\Delta\widetilde{k}=0$. The parameter is the seeded third-harmonic intensity. With filled circles are shown necessary h_{eg}^2 for stationary copropagation of the fundamental and third-harmonic wave.

IV. HIGH-INTENSITY FUNDAMENTAL BEAM

In this section we will use numerical solution of Eq. (1) in order to evaluate the amplitude and phase of the fundamental beam at the output of the nonlinear media. However, some idea of the behavior of the amplitudes can be achieved by the phase portrait technique [19,21,35]. This method allows us easily to evaluate the conditions for stationary waves in the media.

For all calculations presented in this section we chose hypothetical $\chi^{(3)}$ media with $\gamma_3>0$ and the following relations between other nonlinear coupling coefficients $\gamma_1=\gamma_3$, $\gamma_2=2\gamma_3$, $\gamma_4=6\gamma_3$, and $\gamma_5=3\gamma_3$. Such relations are valid for isotropic $\chi^{(3)}$ media. Phase-matching conditions for this type of media can be obtained by the quasi-phase-matching technique [22].

A. Evolution of the fundamental amplitudes

From Eqs. (4a) and (5) it can be obtained that

$$\frac{1}{p}\,\frac{dh^2}{d\xi}=\pm 2\sqrt{h^2(1-h^2)^3-\left[\Gamma_0-\Lambda h^2-\frac{\Delta\gamma}{4}\,h^4\right]^2}. \qquad (16)$$

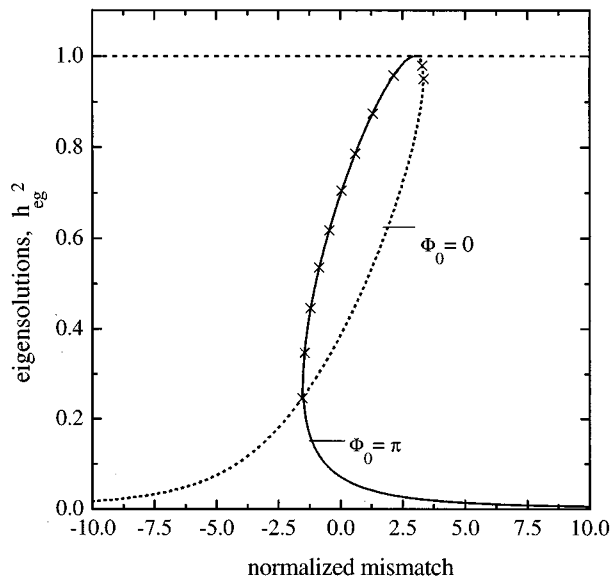

FIG. 5. Third-harmonic eigensolutions vs normalized mismatch $\Delta\widetilde{k}$ for the two input phase differences $\Phi_0=0$ (dotted line) and $\Phi_0=\pi$ (solid line). Trivial eigensolution $h_0^2=1$ is shown with a dashed line. Unstable eigensolutions are marked with crosses ($\times$).

Equation (16) describes trajectories in the phase plane $((1/p)(dh^2/d\xi),h^2)$ that represent the dynamics of the generation of the third-harmonic wave. Lets first consider the case of no third-harmonic signal of the input of the crystal. On Fig. 2 the correspondent curves for different values of the normalized mismatch $\Delta\widetilde{k}=\Delta k/\gamma_3 u^2$ are shown. Closed orbits correspond to periodical energy exchange between the fundamental and the third-harmonic waves. It is interesting to note that only one parameter, $\Delta\widetilde{k}$, is enough to define the amount of the periodic depletion of the fundamental wave, as shown in Fig. 3. Note that the conversion into third harmonic can be maximum 83% when $\Delta\widetilde{k}=-0.56$. In this figure we marked the area where the LD approximation developed in the previous section is appropriate to be used.

For the case of seeded third-harmonic signal the trajectories depend also on the amount of seeding and on the input phase difference between the two input signals Φ_0. The input phase of the seeded wave has strong influence on the process. For $\Phi_0=0,\pi$ eigenmode solutions of the system (4) are possible. Figure 4 represents the phase curves for $\Delta\widetilde{k}=0$ $\Phi_0=0,\pi$ and different values of h_0. From the figure it can be seen that for these initial conditions the propagation of stationary waves (waves that do not alter their intensity but only change their phases) through the nonlinear media is possible, as it is possible in quadratic media [19,20,35].

B. Stationary waves

The stationary waves are characterized by constant amplitude and as follows from Eq. (4a) they can exist only for $\sin\Phi=0$ and, i.e., for $d\Phi/d\xi=0$. Using this condition and Eqs. (4b) and (4c) one may obtain the following equation for the eigensolutions of system (4):

$$[(\Delta\gamma^2+16)h^6+(4\Lambda\Delta\gamma-24)h^4+(4\Lambda^2+9)h^2-1]=0. \tag{17}$$

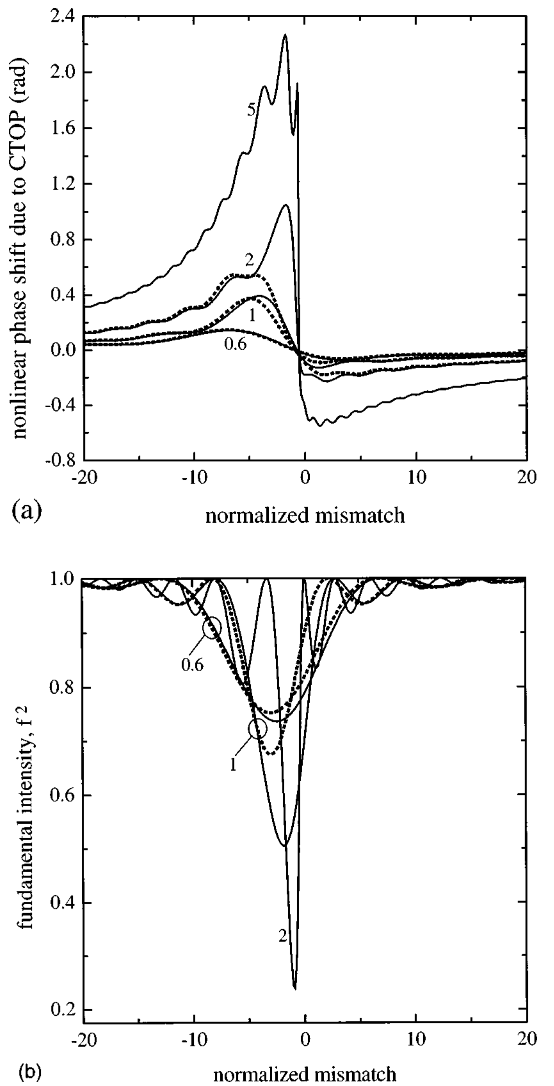

FIG. 6. Cascaded NPS (a) and fundamental intensity (b) vs normalized mismatch $\Delta\widetilde{k}$ for different values of the parameter $p=\gamma_3 u^2 L$ for the case of unseeded third-harmonic generation. The solid line represents the numerical solution of system (1), the dotted line represent the analytical solution obtained from expression (11).

In addition to the trivial solution $h^2=1$, Eq. (17) gives the other eigenmodes of the system (1). In Fig. 5 the dependence of the eigensolutions $h_{\rm eg}^2$ on the normalized mismatch $\Delta\widetilde{k}=\Delta k/\gamma_3 u^2$ is shown for $\Phi_0=0$ and $\Phi_0=\pi$. Some of the eigensolution values do not correspond to the stationary waves in the media and they are marked with ($\times$).

The evolution of the phase for the stationary waves is linear with the propagation distance and the irradiance. Integrating Eq. (4b) we have

$$\varphi_1=\varphi_{10}+[\widetilde{\gamma}_1(1-h_{\rm eg}^2)+\widetilde{\gamma}_2 h_{\rm eg}^2 +\mathrm{sgn}(\cos\Phi_0)\sqrt{(1-h_{\rm eg}^2)}h_{\rm eg}]\gamma_3 u^2 z. \tag{18}$$

Only the last term in the square brackets is connected with the process of third-harmonic generation. We have to bear in mind that in the case of second-harmonic generation the phase evolution of the eigensolutions is linear with the field [19]. In the case of eigensolutions in the third-harmonic

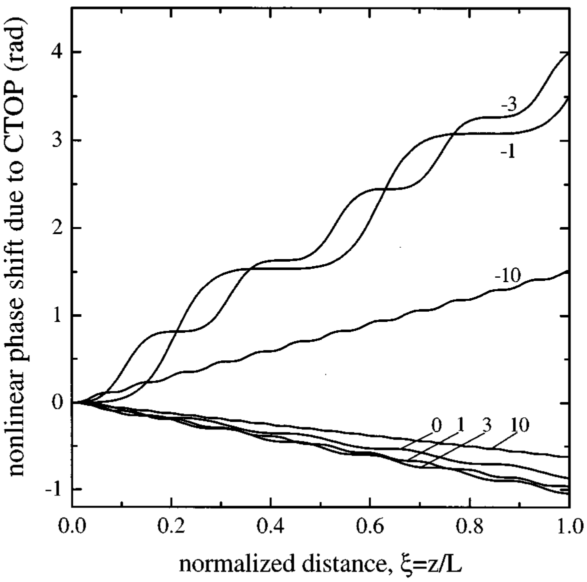

FIG. 7. Variation of the cascaded nonlinear phase shift with the normalized distance $\xi = z/L$ for various phase mismatches for the case of unseeded third-harmonic generation. Normalized input intensity $\gamma_3 u^2 L = 10$.

generation process the nonlinear media acts as lossless media for both waves with the nonlinear index of refraction defined as

$$n_{2,\mathrm{eg}} = n_2 \left[1 + (\gamma_2/\gamma_1) \frac{h_{\mathrm{eg}}^2}{f_{\mathrm{eg}}^2} + (\gamma_3/\gamma_1) \mathrm{sgn}(\cos\Phi_0) \frac{h_{\mathrm{eg}}}{f_{\mathrm{eg}}} \right]. \tag{19}$$

C. The evolution of the fundamental phase

The phase evolution of the fundamental wave can be of primary interest for all optical applications as well as for measurement purposes. For finding the phase we applied direct integration of Eq. (1). The two invariants [Figs. 5(a) and 5(b)] of the system were used to control the accuracy of the calculated data.

Let us first consider the case of zero third-harmonic signal at the input of the nonlinear media. The dependence of the NPS due to CTOP on the normalized mismatch $\Delta\tilde{k}$ is shown in Figs. 6(a) and 6(b). Only the cascaded part of the NPS $\Delta\varphi_{1,\mathrm{casc}} = \Delta\varphi_1 - \gamma_1 u^2 L = \Delta\varphi_1 - p$ is presented on the graph. It can be seen that the dispersionlike curves are centered not at $\Delta\tilde{k} = 0$, but at a small negative value for $\Delta\tilde{k}$ and this can be explained by the correct accounting for the nonlinear refractive indices. For $p \ll 1$, the positive and negative branches of the curves have the same magnitude, while for bigger values of p the positive branch has a higher amplitude (when $\gamma_3 > 0$). The amount of the phase shift due to CTOP is comparable to the phase shift $\Delta\varphi_{\mathrm{dir}}$ due to the inherent $\chi^{(3)}$ of the media ($\Delta\varphi_{1,\mathrm{dir}} = p$). In fact we see that for $p > 1$ the additional phase shift is $\Delta\varphi_{\mathrm{casc}} = (0.4 - 0.5)\Delta\varphi_{\mathrm{dir}}$.

In Fig. 7 the cascaded part of the NPS, $\Delta\varphi_1^{\mathrm{casc}}$ is shown as a function of the normalized length of the media. The normalized mismatch $\Delta\tilde{k}$ was used as a parameter. The phase shift depends linearly on the normalized length ξ for almost all values of $\Delta\tilde{k}$ except for the values between -0.56 and -5, for which high conversion coefficients in the third-harmonic wave are predicted (see Fig. 3). For these values of the $\Delta\tilde{k}$ the NPS due CTOP has a stepwise dependence. The asymmetric behavior of the curves with respect to the normalized phase mismatch ΔkL is evident. For the chosen positive sign of γ_3 and the relations between nonlinear coupling coefficients it is possible to obtain a large positive additional phase shift for negative mismatches.

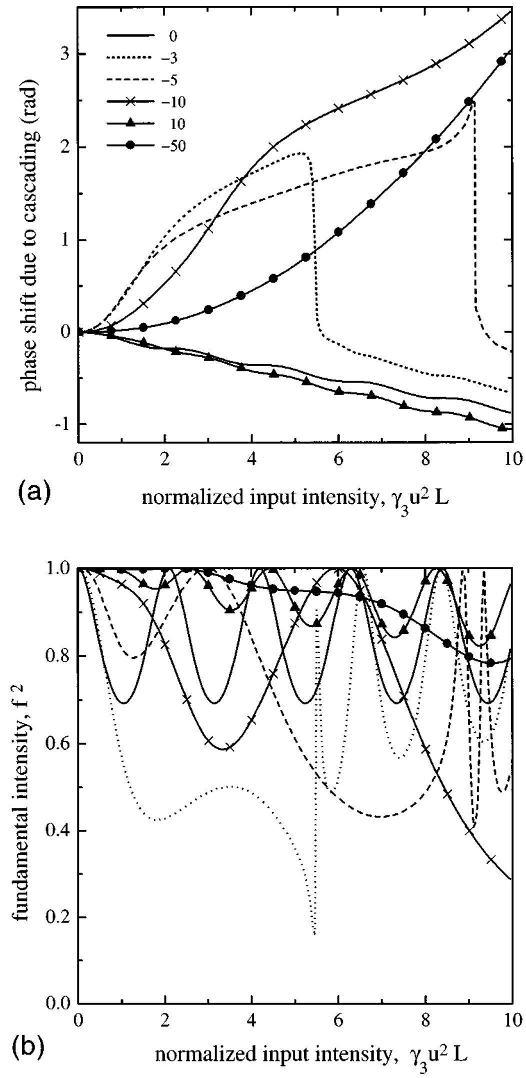

FIG. 8. Cascaded NPS (a) and fundamental transmittance (b) vs the normalized input intensity for the case of unseeded third-harmonic generation. The parameter is the mismatch ΔkL.

The dependence of the cascaded NPS on the input intensity is shown in Fig. 8. The parameter is the normalized ΔkL. Three types of graphs can be distinguished from the figure. The first one is obtained for positive not very big values for the mismatch ($\Delta kL \leq p$). Cascaded NPS is linear with the input intensity. The linear dependence of the NPS due to CTOP on the distance and the input intensity means that the cascade $\chi^{(3)}:\chi^{(3)}$ process has n_2 behavior. For small negative values of ΔkL the depletion of the wave intensity is substantial [Fig. 8(b)] and the cascaded NPS has steplike dependence on the intensity. The jumps in the curves for $\Delta kL = -3$ and $\Delta kL = -5$ correspond to the points where

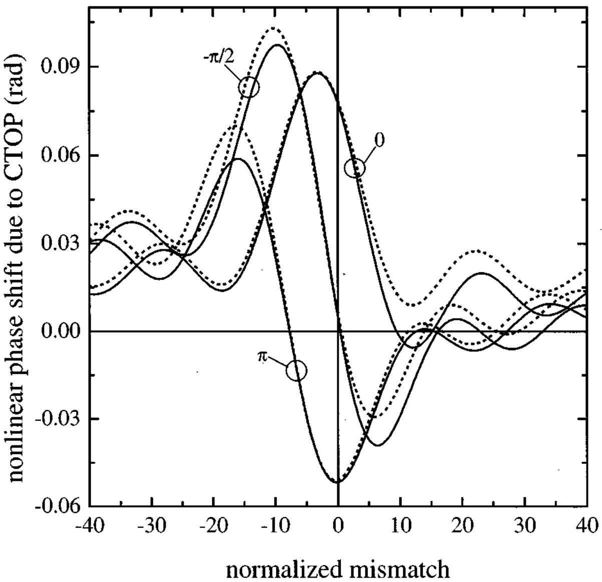

FIG. 9. Cascaded NPS for the case of seeded third-harmonic generation as a function of the normalized mismatch $\Delta\widetilde{k}$. The input phase difference $\Phi_0 = 3\varphi_3(0) - \varphi_1(0)$ is indicated. The normalized input intensity is $\gamma_3 u^2 L = 0.3$. The amount of the seeding is $h_0^2 = 0.05$. The dotted line represents the analytical calculations obtained from Eq. (10). The solid line is the numerical solution of system (1).

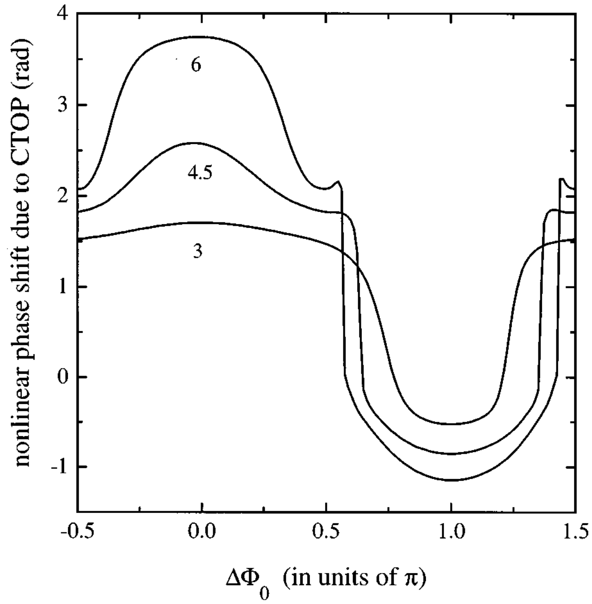

FIG. 10. Cascaded NPS for the case of seeded third-harmonic generation as a function of the input phase difference Φ_0. The normalized input intensity $p = \gamma_3 u^2 L$ is indicated on the graph. The other parameters are the mismatch $\Delta k L = -0.3$ and the seeded intensity $h_0^2 = 0.03$.

the parameter $\Delta\widetilde{k}$ crosses the value -0.56. These abrupt changes of $\Delta\varphi_{\rm casc}$ can be used for constructing all optical switching elements that discriminate the intensity of the input pulses. Quadratic dependence for the NPS as a function of the intensity is obtained for high values of the mismatch, $|\Delta k| L \gg p$ (see the graph for $\Delta k L = -50$). For the range of the intensities shown on the graph the normalized mismatch is $|\Delta\widetilde{k}| \gg 1$ and as we explained in Sec. III B, $\Delta\varphi_{1,\rm casc} \propto \gamma_3^2 u^4 L$. Note that at these conditions the high-value cascaded NPS is combined with relatively low depletion of the fundamental wave intensity.

In the case of the nonzero third-harmonic wave at the input of the media the situation becomes considerably more complex. The relative input phase $\Phi_0 = \varphi_3(0) - 3\varphi_1(0)$ between the seeded and the fundamental beam influences both the magnitude and the collected phase of the interacting waves. We present here two dependencies. The dependence of the cascaded NPS on $\Delta\widetilde{k}$, for $\Phi_0 = 0$, $\Phi_0 = \pi$, and $\Phi_0 = -\pi/2$ is shown in Fig. 9. The change of the input phase leads to a shift of the curves with respect to the nonseeded position (see Fig. 6). We use this graph to demonstrate the applicability of the developed low depletion analytical approach in the previous section.

At input intensities that correspond to $p > 4$ it is possible to obtain a change in the value of the cascaded phase shift by more than π by controlling the input phase difference Φ_0. This is illustrated in Fig. 10, where the cascaded part of NPS is plotted vs the input phase difference Φ_0 for three different values of the parameter p. The seeded third harmonic intensity is 3% of the total input intensity. The change in Φ_0 by π leads to a change in $\Delta\varphi_{\rm casc}$ by values exceeding π. This can be used for constructing switching interferometric devices. If one takes 1-cm-long polydiacetilene paratoluene sulfonate (PTS), as nonlinear media [4,36], then $p = 4.5$ corresponds to 50 MW/cm^2.

V. CONCLUSION

We have investigated the nonlinear phase shift due to two-step cascaded third-order processes in centrosymmetric media at different input conditions. NPS due to CTOP can be of the same order of magnitude as NPS due to direct $\chi^{(3)}$ processes.

For low input intensities ($p \ll |\Delta k| L$) the NPS due to CTOP has n_4 behavior (cascaded NPS is linear with distance and quadratic with input intensity). For higher input intensities ($p \gg |\Delta k| L$) NPS due to CTOP has a behavior that is closer to n_2 (linear with distance and input intensity after averaging over the steps). This means that the nonlinear phase shift due to $\chi^{(3)}:\chi^{(3)}$ processes has a similar behavior to the nonlinear phase shift due to $\chi^{(2)}:\chi^{(2)}$ processes only in the sense that there is a saturation with increasing irradiance. We remind that the initial dependence of the NPS due to $\chi^{(2)}:\chi^{(2)}$ processes on the field is quadratic and at higher irradiance becomes linear.

It is interesting to perform similar analysis for type-II phase-matched third-harmonic generation. As in the case of type-II phase second-harmonic generation [37,38], but now as a result of cascade $\chi^{(3)}:\chi^{(3)}$ processes, cross phase modulation for the two fundamental waves is observed [39]. The third-order cascade processes should influence the phase of the pump beams involved in other types phase matchable cubic processes as four-wave mixing, electric-field-induced second-harmonic generation and phase conjugation.

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