Cross-phase modulation caused by cascading of third-order processes

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The evolution of the phase of a weak signal wave (ω_2) copropagating with a strong gate wave (ω_1) in a centrosymmetric medium is studied analytically and numerically for the case when the two waves are involved in a nearly phase-matched third-order process that generates a third wave ($\omega_3 = 2\omega_1 + \omega_2$). It is shown that the signal wave collects, in addition to the phase shift that is due to the $\omega_2 = \omega_1 + \omega_2 - \omega_1$ process, an extra nonlinear phase shift caused by cascading of two third-order nonlinear processes, $\omega_3 = 2\omega_1 + \omega_2$ and $\omega_2 = \omega_3 - 2\omega_1$. The attractive feature of this new type of cross-phase modulation is the fact that one can control its magnitude and sign by changing the wave-vector mismatch of the sum-frequency mixing process and the gate intensity. [S0740-3224(99)01702-6]

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1. INTRODUCTION

Cross-phase modulation (XPM) induced by coupling among optical waves gives rise to a number of important nonlinear effects[1] such as induced nonlinear birefringence and the associated optical Kerr effect, polarization instability, and pulse shaping; spectral broadening and generation of a continuum, modulation instability, and a solitary wave supported by coupling through XPM[2,3]; and weak-beam deflection in an off-axis geometry.[4] The two-color Z-scan technique used for measurements of $\chi^{(3)}$ nondegenerate components[5–7] and laser pulse duration[8] is also based on the effects of XPM. The possibility of maximizing the effects of XPM to control its magnitude and sign will be useful in the future developments of these applications.

Recently it was shown (Refs. 9–12 and references therein) both theoretically and experimentally that in a noncentrosymmetric medium tuned for nearly phase-matched type II second-harmonic generation or sum-frequency mixing it is possible to achieve the XPM effect by use of cascaded second-order processes. The effect is described by an artificial cubic nonlinearity $\chi^{(3)}_{\text{casc}} \propto [\chi^{(2)}]^2$, which as a rule has a higher value than the inherent cubic nonlinearity of the medium. Importantly, one can change the sign of $\chi^{(3)}_{\text{casc}}$ simply by reversing the sign of the wave-vector mismatch of the process. In centrosymmetric media $\chi^{(2)} = 0$, and the second-order cascade processes cannot be used to enhance or reverse the sign of the XPM effect. However, one can expect that in centrosymmetric media cascaded third-order processes[13,14] will behave similarly. As far as we know, this problem has not been addressed.

Here we show theoretically that, under certain conditions, cascaded third-order processing (CTOP) can yield a XPM effect comparable in magnitude with the XPM that is due to intrinsic $\chi^{(3)}$ of the medium. By changing the sign of this cascaded XPM it is possible to achieve enhancement or reduction of the overall XPM effects.

2. PLANE-WAVE EQUATIONS

We consider two linearly polarized plane waves, E_1 and E_2, copropagating through a centrosymmetric lossless medium. The two waves interact throughout the cubic nonlinearity of the media, generating a third wave, E_3. The total field in the medium is taken to be

$$E(z, t) = \frac{1}{2}\sum e_j A_j(z)\exp[i(\omega_j t - k_j z)] + \text{c.c.},$$

where $k_j = 2\pi n_j/\lambda_j$ and ω_j are, respectively, the propagation constants and frequencies. A_j are the complex slowly varying wave amplitudes, which 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 relevant slowly varying envelope equations for the process $2\omega_1 + \omega_2 = \omega_3$ have the form

$$\frac{dA_1}{dz} = iW_1 A_1 + i\gamma_1 A_3 A_2^* A_1^* \exp(i\Delta kz),$$

$$\frac{dA_2}{dz} = iW_2 A_2 + i\gamma_2 A_3 A_1^* A_1^* \exp(i\Delta kz),$$

$$\frac{dA_3}{dz} = iW_3 A_3 + i\gamma_3 A_1 A_1 A_2 \exp(-i\Delta kz), \tag{1}$$

where $W_j = \Sigma_l \gamma_{jl} a_l^2$ ($j, l = 1, 2, 3$) and the wave-vector mismatch is $\Delta k = k_3 - 2k_1 - k_2$. In the case of parallel polarization of the input waves the nonlinear coupling coefficients γ_j and γ_{jl} obey the following relations: $\gamma_j = (\omega_j/2\omega_1)\gamma_1$, $j = 2,3$; $\gamma_{jl} = (\omega_j/\omega_1)(2 - \delta_{jl})\gamma_0$, $j,l = 1, 2, 3$; and $\gamma_0 = (3\pi/4\lambda_1 n_1)\chi^{(3)}$.

Equations (1) include only those four-wave interactions that have relatively small wave-vector mismatches ($\Delta k < 50/L$, where L is the length of the nonlinear medium). Other four-wave interactions, including those that yield the third harmonic of the input waves and these that are responsible for optical rectification, are considered to be far from the phase-matching condition and inefficient (see the discussion in Section 5 below).

When ω_2, k_1, k_2, and k_3 are replaced with ω_1, $k_1^{(o)}$, $k_1^{(e)}$, and $k_3^{(e)}$, respectively, in Eqs. (1), those equations also describe type II third-harmonic generation, i.e., one ordinary and one extraordinary wave generate an extraordinary third-harmonic wave. The wave-vector mismatch is then $\Delta k = k_{3\omega}^{(e)} - 2k_{1\omega}^{(o)} - k_{1\omega}^{(e)}$. However, in this case the relationships between the nonlinear coupling coefficients will be different.

In principle Eqs. (1) can be integrated exactly. The resultant analytical expressions for the amplitude and the phase of the interacting waves consist of Jacoby elliptical functions and integrals; however, the use of these analytical solutions is rather difficult because evaluation of the elliptical sine and the elliptical integral of the third kind (used in the expressions for the nonlinear phase shift) requires complicated numerical calculations.[15] Thus to solve Eqs. (1) we chose direct numerical integration and derivation of an approximate analytical formula by using a low-depletion approximation.[14,16]

In what follows, we shall consider the situation when $|a_2|^2 \ll |a_1|^2$, calling these two fundamental waves signal and gate waves, respectively. The case of equal intensity of the two fundamental waves is practically similar to a case of third-harmonic generation that has already been considered.[13,14]

3. ANALYTICAL APPROACH

The system of Eqs. (1), when it is rewritten with respect to the real amplitudes and the phases, has the following form:

$$a_j \frac{\mathrm{d}a_j}{\mathrm{d}z} = \pm\gamma_j a_1^2 a_2 a_3 \sin\Phi, \tag{2a}$$

$$a_j \frac{\mathrm{d}\varphi_j}{\mathrm{d}z} = W_j a_j + \frac{\gamma_j a_1^2 a_2 a_3}{a_j} \cos\Phi, \tag{2b}$$

where $j = 1,2,3$ and $\Phi = \varphi_3 - 2\varphi_1 - \varphi_2 + \Delta kz$. The minus in Eq. (2a) corresponds to $j = 1,2$, and the plus corresponds to $j = 3$.

With the suggestions that $a_3(0) = 0$, $a_1(0) = a_{10}$, and $a_2(0) = a_{20}$, the three invariants of Eqs. (2) are

$$a_j^2 + \frac{\gamma_j}{\gamma_3} a_3^2 = a_{j0}^2, \qquad j = 1,2, \tag{3}$$

$$\gamma_3 a_1^2 a_2 a_3 \cos\Phi + \Lambda a_3^2 + \frac{\Delta\gamma}{4} a_3^4 = 0. \tag{4}$$

In Eq. (4) the following substitutions have been made:

$$\Lambda = (\Delta k + \Gamma_1 a_{10}^2 + \Gamma_2 a_{20}^2)/2,$$

$$\Delta\gamma = (\Gamma_3\gamma_3 - \Gamma_1\gamma_1 - \Gamma_2\gamma_2)/\gamma_3,$$

$$\Gamma_j = -2\gamma_{1j} - \gamma_{2j} + \gamma_{3j} \qquad (j = 1, 2, 3).$$

Approximate expressions for the square of the amplitude of the interacting waves can be derived[14,16] for the situation when the coefficient of the intensity conversion into the third wave with respect to the gate wave does not exceed 20%:

$$a_j^2(z) = a_{j0}^2 - \frac{\gamma_j}{\gamma_3} a_3^2(z) \qquad (j = 1, 2), \tag{5}$$

$$a_3^2(z) = \frac{\gamma_3^2}{Q^2} a_{10}^4 a_{20}^2 \sin^2(Qz), \tag{6}$$

where $Q^2 = \gamma_3(2\gamma_1 a_{10}^2 a_{20}^2 + \gamma_2 a_{10}^4) + \Lambda^2$.

The phase of the weak wave can be obtained by integration of Eq. (2b) for $j = 2$. Two different approaches are possible: weak depletion of the signal intensity and strong periodic depletion of the signal intensity.

In the first case the phase shift $\Delta\varphi_2$ of the signal wave at the output of the crystal has the form

$$\Delta\varphi_2 \approx \gamma_{21} a_{10}^2 L - \frac{\gamma_3\gamma_2 a_{10}^4 L}{2\Lambda}[1 - \mathrm{sinc}(2\Lambda L)]. \tag{7}$$

This expression is useful for interpreting the physics of the XPM that are due to CTOP. In the second case (strong periodic depletion of the signal intensity), as a result of exact integration of Eq. (2b), $j = 2$ we obtained a more complicated but more accurate expression for $\Delta\varphi_2$:

$$\Delta\varphi_2 = \left(A + \gamma_3^2 \frac{B a_{10}^4 a_{20}^2}{2Q^2}\right) L - \gamma_3^2 \frac{B a_{10}^4 a_{20}^2 L}{2Q^2} \frac{\sin(2QL)}{2QL} - \frac{4\gamma_2\Lambda + \gamma_3\Delta\gamma a_{20}^2}{4\gamma_2 QD} \arctan[D\tan(QL)], \tag{8}$$

where

$$A = \gamma_{21} a_{10}^2 + \left(\gamma_{22} + \frac{\gamma_3\Delta\gamma}{4\gamma_2}\right) a_{20}^2 + \Lambda,$$

$$B = \gamma_{23} - \frac{\gamma_{21}\gamma_1}{\gamma_3} - \frac{\gamma_{22}\gamma_2}{\gamma_3} + \frac{\Delta\gamma}{4},$$

$$D = \left(1 - \frac{\gamma_2\gamma_3 a_{10}^4}{Q^2}\right)^{1/2}.$$

4. PHYSICAL INTERPRETATION

A physical explanation of the effect of XPM that is due to CTOP can be obtained by consideration of relation (7). The first term represents the phase shift of the signal wave that is the contribution of the single third-order process $\omega_2 = \omega_1 + \omega_2 - \omega_1$ known as XPM. This process is always phase matched and does not depend on Δk.

The second term accounts for the XPM that is a result of cascading of two third-order processes:

(i) Generation of a third wave with frequency $\omega_3 = 2\omega_1 + \omega_2$; during this process the weak wave is depleted.

(ii) Reconstruction of the intensity of the signal wave through the process $\omega_2 = \omega_3 - 2\omega_1$.

As can be seen from relation (7), this additional term depends on the square of intensity of the gate wave, $I_1^2 \propto a_{10}^4$, indicating directly that it represents high-order XPM effects. From the other side the second term in relation (7) is proportional to the product of two nonlinearities, $\gamma_2\gamma_3 \propto \chi^{(3)}\chi^{(3)}$. This is the usual signature of the cascading processes that was used to introduce the terminology $\chi^{(2)}:\chi^{(2)}$ cascading for cascading of second-order processes[12,17] and $\chi^{(3)}:\chi^{(3)}$ cascading[13,14] in our case. The term that is responsible for XPM that is due to CTOP becomes 0 when $\Lambda = 0$. Within the suggestion that $a_{20}^2 \ll a_{10}^2$ this corresponds to the condition $(\Delta k/\gamma_0 a_{10}^2) = \Gamma_1/\gamma_0$. The applicability of relation (7) is for $\Delta\tilde{k} = (\Delta k/\gamma_0 a_{10}^2) \gg 1$. As we shall see below, for lower values of $\Delta\tilde{k}$ (i.e., higher values of $\gamma_0 a_{10}^2 L$) the cascaded part of the nonlinear phase shift is no longer proportional to I_1^2.

5. RESULTS AND DISCUSSIONS

For calculations with the more-general Eq. (8) and for numerical integration of Eqs. (1) we chose a hypothetical experiment for which the signal wave is the second harmonic of the gate wave, $\omega_2 = 2\omega_1$, the ratio of input intensities is $a_{20}^2/a_{10}^2 = 0.01$, and the ratio of the two nonlinear coupling coefficients is $\gamma_1/\gamma_0 = 2$. The generated third wave will have the frequency $\omega_3 = 4\omega_1$.

The dependence of the signal wave's nonlinear phase shift on the normalized mismatch $\Delta\tilde{k}$ is shown in Fig. 1, where only cascaded part of the nonlinear phase shift $\Delta\varphi_{2,\mathrm{casc}} = \Delta\varphi_2 - (\gamma_{21} a_{10}^2 - \gamma_{22} a_{10}^2)L$ is shown. The curves are centered at $\tilde{k} = -2$, close to the value predicted from relation (7). For comparison, self-phase modulation that is due to CTOP of the fundamental wave involved in type I third-harmonic generation[14] is shown by a dashed curve. This is the case when the two input waves are indistinguishable ($\omega_1 = \omega_2$, $a_{10}^2 = a_{20}^2$). It can be seen that the effect of XPM that is due to cascading is very strong in comparison with the effect of self-phase modulation that is due to $\chi^{(3)}:\chi^{(3)}$ cascading and comparable with the XPM that is due to natural $\chi^{(3)}$ of the medium that is equal to $\gamma_{21} a_{10}^2 L$. Figure 1 demonstrates the good agreement between numerical results and the analytical formula obtained with the low-depletion approach.

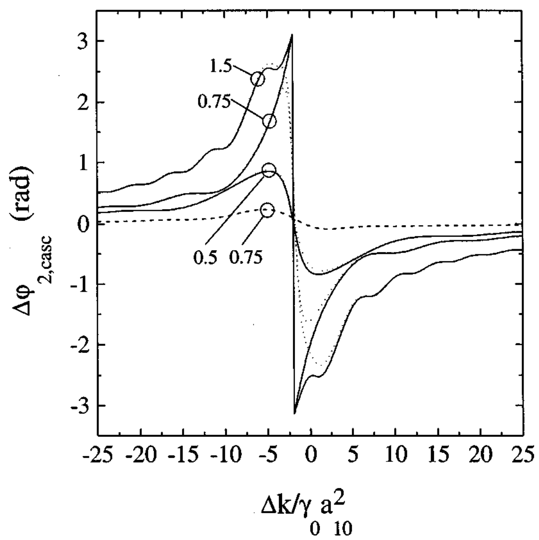

Fig. 1. Cascaded NPS $\Delta\varphi_{2,\mathrm{casc}}$ as a function of the normalized mismatch $\Delta\tilde{k}$ for several values of the normalized gate intensity $\gamma_0 a_{10}^2 L$. Solid curves, numerical solution of Eqs. (1); dotted curves, analytical solution obtained from Eq. (8). The ratio of the signal and the gate input intensities is $a_{20}^2/a_{10}^2 = 0.01$. Dashed curve, NPS that is due to CTOP when $\omega_1 = \omega_2$ and $a_{10}^2 = a_{20}^2$ (this curve is reproduced from Ref. 14).

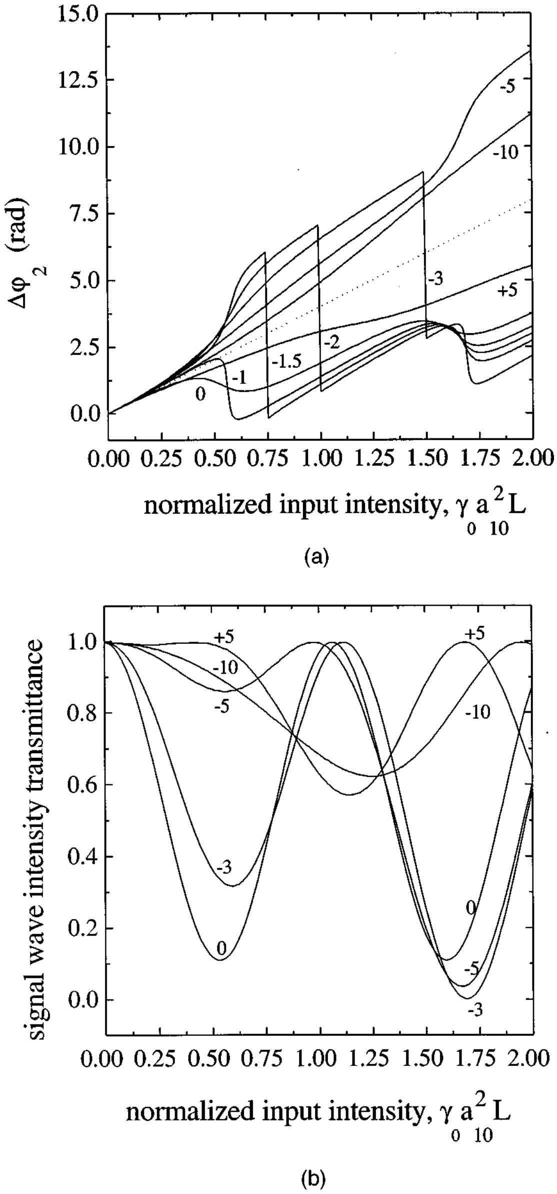

Fig. 2. Variation of (a) total NPS $\Delta\varphi_2$ and (b) signal intensity transmittance a_2^2/a_{20}^2 with the normalized gate intensity. The parameter is the mismatch ΔkL. Dotted line, NPS that is due to pure XPM. The ratio of the signal and gate input intensities is $a_{20}^2/a_{10}^2 = 0.01$.

In Fig. 2 the total nonlinear phase shift (NPS) and intensity of the signal wave are shown as functions of the normalized input intensity $\gamma_0 a_{10}^2 L$. The parameter is the normalized mismatch ΔkL. The dotted line represents the NPS that is due to pure, single-step XPM. The figure clearly shows that by using CTOP one can double or substantially reduce the XPM effect, depending on the mismatch ΔkL. We recall that the magnitude of the self-phase modulation that is due to CTOP reaches a maximum of 30% of the self-phase modulation caused by Kerr nonlinearity of the medium.[14] The 2π jumps in the curves correspond to the points where the parameter $\Delta\tilde{k} = (\Delta k/\gamma_0 a_{10}^2)$ crosses the value -2. From the figure it

Table 1. Values of the Wave-Vector Mismatches (in cm^{-1}) for Different Processes in Lead Molybdate Crystal[a]

Variable	New Frequency Generation Process				Optical Rectification Process	
Δk	$k_{4\omega}^{(e)} - 2k_{1\omega}^{(o)} - k_{2\omega}^{(o)}$	$k_{3\omega}^{(e)} - 2k_{1\omega}^{(o)} - k_{1\omega}^{(e)}$	$k_{3\omega}^{(e)} - 3k_{1\omega}^{(o)}$	$k_{3\omega}^{(e)} - 2k_{2\omega}^{(o)} + k_{1\omega}^{(o)}$	$k_{4\omega}^{(e)} - 2k_{2\omega}^{(o)}$	$k_{2\omega}^{(o)} - 2k_{1\omega}^{(o)}$
Value	8.7	−477	−2200	−4011	−897.0	906

[a] The angle between the direction of the input waves and the Z axis is 62°.

can also be seen that for higher values of input intensity when $\Delta\tilde{k} < 1$ the dependence of the NPS on intensity is no longer quadratic with respect to the intensity of the gate wave. So the effect of XPM that is due to cascading can be considered a result of effective quintic nonlinearity only for low input gate intensities $\Delta\tilde{k} > 1$. Figure 2(b) reveals that the strong XPM effect is accompanied by periodic depletion and reconstruction of the signal wave. To avoid the losses for the signal wave one should work at the points of full reconstruction of the signal wave.

Significant XPM owing to cascading can be observed at a normalized input intensity as low as $\gamma_0 a_{10}^2 L = 1$, which corresponds to $I_1 \approx 23\,\mathrm{MW/cm^2}$ if one takes as the medium 1-cm long *p*-toluene sulfonate (PTS),[18,19] $\lambda_1 = 2.62\,\mu\mathrm{m}$ for the gate wavelength, and $\lambda_2 = 1.31\,\mu\mathrm{m}$ for the signal wavelength. These levels of power density can easily be achieved with many pulse sources. We took the crystal PTS for this estimation because it has the largest known cubic nonlinearity [$\chi^{(3)} = 1.6 \times 10^{-10}$ esu (Ref. 18)] and is transparent for all interacting waves when the gate wave is at $\lambda_1 = 2.62\,\mu\mathrm{m}$. Reference 18 reports third-harmonic generation in PTS of a fundamental beam at this wavelength and strong four-wave parametric amplification. The lack of published data for the dispersion of the refractive index of this crystal does not allow us to predict possible phase matching, but judging from its very high value of birefringence, $n_{zz} - n_{xx} = 0.3$,[18] it is extremely likely that PTS permits birefringent phase matching.

Another medium suitable for observation of cascaded XPM is lead molybdate crystal ($PbMoO_4$). This crystal has a relatively high third-order susceptibility, $\chi_{xxxx}^{(3)} \times (PbMoO_4) = 1.35\chi_{xxxx}^{(3)}(CS_2)$,[20,21] and a dispersion of the refractive index[22] such that phase matching is possible not only for four-wave upconversion ($\omega_1, \omega_1, 2\omega_1, 4\omega_1$) with $\lambda_1 = 2.62\,\mu\mathrm{m}$ but also for another type of four-wave interaction for which all three input waves have the same frequency, but the polarization of the weak signal wave is perpendicular to that of the gate wave. The latter interaction is usually called type II third-harmonic generation and, as we have already mentioned, is also described by Eqs. (1).

We used lead molybdate crystal to calculate the relative magnitude of the wave-vector mismatches for different four-wave interactions when one of the processes is phase matched. The exact phase-matching angle for the process considered by us, $k_{4\omega}^{(e)} = 2k_{1\omega}^{(o)} + k_{2\omega}^{(o)}$, is $\theta = 62.06°$. The mismatches for the different types of interaction are shown in Table 1. The processes with the high-value mismatches will be inefficient, and for this reason they have not been included in Eqs. (1).

For media that do not possess the necessary birefringence for angle-tuning phase matching, the so called quasi-phase-matching technique[23] can be applied. Recently this method was further developed, and it now can be applied for achieving exact or near phase matching for four-wave interactions in centrosymmetric media.[24,25] The authors of Ref. 24 used a multilayer stack made from glass coverslips covered with highly nonlinear film to obtain third-harmonic generation from a radiation at 1.542 μm. The second type of quasi-phase-matching technique that is suitable to be applied for centrosymmetric media uses spatial periodic modulation of the fundamental wave intensity along the propagation direction without artificial modulation of the nonlinear optical properties.[25]

Another way to achieve near phase matching in centrosymmetric media is to use mixtures of gases[26] or solvents.[27,28]

6. CONCLUSION

In conclusion, we have described a new type of cross-phase matching that is due to cascaded third-order processes in media with inversion centers. This way to generate an induced nonlinear phase shift is a high-order analog to the induced nonlinear phase shift that is due to the second-order cascading[9–12] observed during sum-frequency mixing interactions in noncentrosymmetric media. The results presented here confirm that the process of generating a NPS in nondegenerate frequency mixing interactions is more efficient than the interactions that use type I harmonic generation, i.e., when the input waves are indistinguishable.

We believe that the strong non-Kerr XPM effect described in this paper can play an important role in analyzing the existence and stability of bright and dark solitary waves supported by the process of nearly phase-matched sum-frequency generation, similarly as for the case of nearly phase-matched third-harmonic generation.[29,30]

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