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ANALYTICAL AND NUMERICAL INVESTIGATION OF OPTO-OPTICAL PHASE MODULATION BASED ON COUPLED SECOND ORDER NONLINEAR PROCESSES

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Abstract. It is shown that the induced phase shift gained by any of the fundamental waves in such processes as type II second harmonic generation and sum frequency mixing depends on the intensity of both input waves. This provides a means of control of the phase shifts that is essential for construction of ultra fast all-optical switching gates. The induced phase shift is a result of simultaneous action of coupled second order processes and is described by effective cubic non linearity.

1. Introduction

The influence of second order processes on the efficiency of the third order interactions in noncentrosymmetric media was noticed long ago [1, 2]. In [3] it was shown that the effective cubic susceptibility responsible for phase matched third harmonic generation in noncentrosymmetric crystals is a sum of two terms: intrinsing $\chi^{(3),\mathrm{int}}$ and so called "cascade" third order susceptibility $\chi_{\mathrm{eff}}^{(3),\mathrm{casc}}$. Four and five order processes are also affected by the low order processes [4–6].

The investigation of the role of second order processes on third order nonlinear optical processes have recently received considerable attention [7–12]. It was shown that the effective $\bar{n}_2$ can be 2×10^{-14} cm^2/W for 1 mm KTP crystal [7] and as much as 2×10^{-11} cm^2/W for organic crystals [11]. The large phase shifts caused by this effective $\bar{n}_2$ can prove useful for low power ultra fast all-optical switching [12], Kerr mode-locking [13], pulse compressing [14], formation of solitonlike waves [15] and others.

Up to now only the effect of self-phase modulation via "cascaded" second order processes near phase matching direction for the type I collinear second harmonic generation was considered [7, 9–11, 16]. Some aspects of the cross phase modulation experienced by the fundamental waves for the case of sum frequency modulation are presented in

[17].

Here we provide analytical and numerical analysis of the process of opto-optical phase modulation experienced by the fundamental waves in such second order nonlinear processes as type II second harmonic generation and sum frequency mixing.

2. Theoretical Analysis

We start our investigation of the process of sum frequency mixing $\omega_c = \omega_b + \omega_a$ (Fig. 1) in nonlinear crystal (NLC) with length L. We assume that the three interacting fields are linearly polarized plane waves. The total electric field can be written as

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

where $j = a, b, c$; $A_j(z)$ are the complex amplitudes. They incorporate both the real amplitude and the phase of the j wave: $A_j(z) = a_j(z)\exp[i\varphi_j(z)]$; $k_j = \frac{2\pi}{\lambda_j}n_j$ and ω_j are the corresponding propagating constants and frequencies. e_a, e_b, e_c are the polarization unit vectors of the three waves. In our investigations only second order nonlinear processes are taken into account. The amplitude equations in the slowly-varying envelope approximation, with assumption of zero absorption for all interacting waves, have the following form:

$$\frac{dA_a}{dz} = -i\sigma_a A_c A_b^* \exp(-i\Delta kz) \tag{2}$$

$$\frac{dA_b}{dz} = -i\sigma_b A_c A_a^* \exp(-i\Delta kz) \tag{3}$$

$$\frac{dA_c}{dz} = -i\sigma_c A_a A_b \exp(i\Delta kz) \tag{4}$$

where the subscripts "a" and "b" denote the fundamental waves and the subscript "c" denotes the generated wave. The wave vector mismatch is $\Delta k = k_c - k_b - k_a$. Nonlinear coupling coefficients σ_a, σ_b, σ_c include the contraction of second order susceptibility tenzor $d^{(2)} = \frac{\chi^{(2)}}{2}$ with the polarization unit vectors:

$$\sigma_a = \frac{2\pi}{\lambda_a n_a} d_{\text{eff}} = \frac{2\pi}{\lambda_a n_a}(e_a . d^{(2)} : e_b e_c) \tag{5}$$

$$\sigma_b = \frac{2\pi}{\lambda_b n_b} d_{\text{eff}} = \frac{2\pi}{\lambda_b n_b}(e_b . d^{(2)} : e_a e_c) \tag{6}$$

$$\sigma_c = \frac{2\pi}{\lambda_c n_c} d_{\text{eff}} = \frac{2\pi}{\lambda_c n_c}(e_c . d^{(2)} : e_a e_b)\,. \tag{7}$$

The same equations describe the process of type II second harmonic generation. For this case A_c is the second harmonic field complex amplitude, A_a and A_b — the complex amplitudes of the two ortogonally polarized input waves.

The system (2–4) can be rewritten in this way:

$$\frac{d^2A_a}{dz^2} + i\Delta k\frac{dA_a}{dz} + \sigma_a A_a(\sigma_c|A_b|^2 - \sigma_b|A_c|^2) = 0 \tag{8}$$

$$\frac{d^2A_b}{dz^2} + i\Delta k\frac{dA_b}{dz} + \sigma_b A_b(\sigma_c|A_a|^2 - \sigma_a|A_c|^2) = 0 \tag{9}$$

$$\frac{d^2A_c}{dz^2} + i\Delta k\frac{dA_c}{dz} + \sigma_c A_c(\sigma_a|A_b|^2 - \sigma_b|A_a|^2) = 0\,. \tag{10}$$

For exact solution of the output phases of the fundamental beams the system (8–10) was solved by us numerically, but in order to analyse the physical meaning of the process of cross phase modulation of the two fundamental beams let us first consider the approximation of the fixed intensity for the fundamental beams [16]. In this approximation the real amplitudes a_a and a_b of the fundamental waves are considered not depending on z, but the phases ϕ_a and ϕ_b are functions of z. Denoting the constant $\sigma_c(\sigma_b a_a^2 + \sigma_a a_b^2) = S$, which depends on input intensities and the nonlinear optical properties of the NLC. Solving (10) with $a_c(0) = 0$, we get for the amplitude and the phase of the generated wave

$$a_c(z) = \sigma_c a_a(0) a_b(0) z \operatorname{sinc}\left(\frac{1}{2}Kz\right) \tag{11}$$

$$\varphi_c(z) = \varphi_a(0) + \varphi_b(0) - \frac{\pi}{2} + \frac{\Delta kz}{2} \tag{12}$$

where $K = \sqrt{\Delta k^2 + 4S}$.

For the phases of the interacting waves the following equations can be obtained from (2-4):

$$\frac{d\varphi_a}{dz} = -\sigma_a\frac{a_c a_b}{a_a}\cos\left(\varphi_c - \varphi_a - \varphi_b - \Delta kz\right) \tag{13}$$

$$\frac{d\varphi_b}{dz} = -\sigma_b\frac{a_c a_a}{a_b}\cos\left(\varphi_c - \varphi_a - \varphi_b - \Delta kz\right) \tag{14}$$

$$\frac{d\varphi_c}{dz} = -\sigma_c\frac{a_a a_b}{a_c}\cos\left(\varphi_c - \varphi_a - \varphi_b - \Delta kz\right)\,. \tag{15}$$

From the last equation we obtain that

$$\cos\left(\varphi_c - \varphi_a - \varphi_b - \Delta kz\right) = \frac{\Delta k a_c}{2\sigma_c a_a a_b}\,. \tag{16}$$

Substituting in (13–14) the expression (11) and (16) and after integration we get for the phase shifts at the end of the NLC $\Delta\varphi_j = \varphi_j(L) - \varphi_j(0) (j = a, b)$:

$$\Delta\varphi_a = \sigma_c\sigma_a a_b^2 L^2 \frac{\Delta kL}{(KL)^2}\left[1 - \operatorname{sinc}(KL)\right] \tag{17}$$

$$\Delta\varphi_b = \sigma_c\sigma_b a_a^2 L^2 \frac{\Delta kL}{(KL)^2}\left[1 - \operatorname{sinc}(KL)\right]\,. \tag{18}$$

3. Discussion

As can be seen from (17) and (18) the induced phase shift $\Delta\varphi_a$ depends linearly on the intensity of the fundamental beam "b" and the induced phase shift $\Delta\varphi_b$ depends linearly on the intensity of the fundamental beam "a". Very important fact, confirmed also by the numerical solution, is that $\Delta\varphi_a$ and $\Delta\varphi_b$ do not depend on the input phases $\varphi_a(0)$ and $\varphi_b(0)$, respectively. If we take both input waves to be identical (as it is for type I second harmonic generation), then the expressions (17) and (18) take the form obtained for the first time in [16].

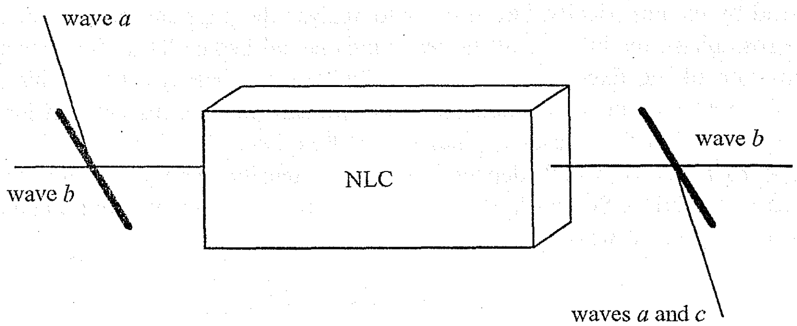


Fig. 1. Schematic representation of quasi phase-matched wave interaction in nonlinear crystal without center of inversion for obtaining optically controlled large induced phase shift: wave "a" is the source wave; wave "b" — the signal wave; wave "c" — the generated wave. The output phase shift of the wave "b" is proportional to the intensity of the wave "a"

It is clear from (17) and (18) that the phases of both fundamental waves experience phase shift, but for our discussions we will consider wave "b" as a signal wave and wave "a" as a source wave, that control the phase of the signal, as shown in Fig. 1. In Fig. 2 are shown the analytical (expression (14)) and the exact numerical solution of the system (8–10) for the phase shift $\Delta\varphi_b$ as a function of normalized phase mismatch ΔkL for four different values of the "normalized total input intensity" SL^2. Input intensities of both input waves are taken to be equal. From this graphs we see that the phase change of the signal wave is described by dispersion-like curve centred around exact phase-matched position $\Delta kL = 0$. Even for the high intensity input beams, where the depletion of the fundamental beams is quite substantial, the analytical graphs are very close to the graphs obtained by the more precise numerical solution of the system (8–10). If the "normalized total input intensity" SL^2 does not exceed 2, maximum phase shift is obtained for normalized mismatches in the range $\Delta kL = (0.5 - 1)\pi$. The exact optimum value $(\Delta kL)_{\text{opt}}$, obtained by numerical solution, depends on the parameter S and on the ratio of the input intensities (Fig. 3). Analytical formulae (17–18) give for the $(\Delta kL)_{\text{opt}}$ a value of π in the validity range of the approximation $\dfrac{2\sqrt{S}}{\Delta k} \ll 1$ (see dashed line in Fig. 3).

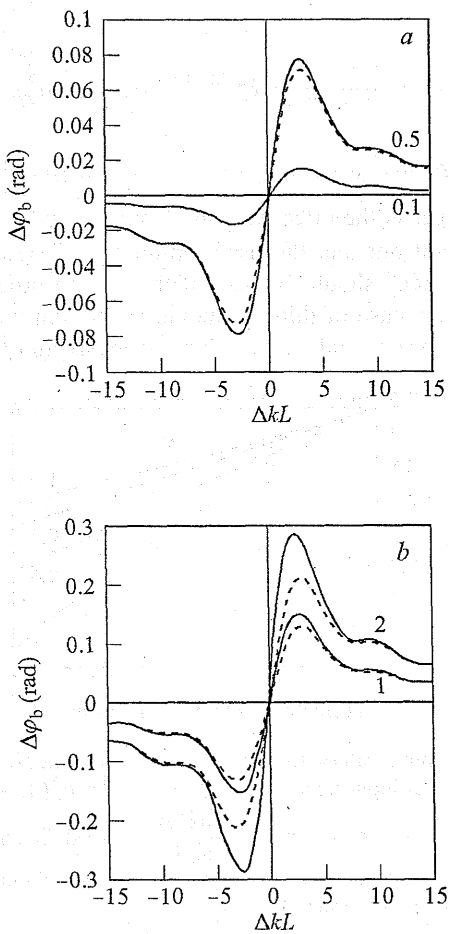


Fig. 2. Output phase shift of the wave "b" as a function of the phase mismatch ΔkL for input intensities that satisfy $\sigma_b I_a = \sigma_a I_b$. Dashed line is the analytical solution obtained from fixed intensity approximation (expression (18)). Solid line is the numerical solution of system (8–10). The parameter is the value of the "normalized total input intensity" $SL^2 = \sigma_c L^2 \left[\sigma_b a_a^2(0) + \sigma_b a_b^2(0)\right]$

In the same range of small input intensities $\dfrac{2\sqrt{S}}{\Delta k} \ll 1$ maximum phase shift is described by:

$$\Delta\varphi_b^{\max} = \frac{\sigma_b \sigma_c L}{\pi} a_a^2 L\,. \tag{19}$$

From the other side for the phase shift $\Delta\varphi_b$ obtained in the process of cross phase modulation ($\omega_b = \omega_a + \omega_b - \omega_a$) due to $\chi^{(3),\,\mathrm{int}}$ only the following expression is valid:

$$\Delta\varphi_b = \frac{\pi}{\lambda_b n_b}\frac{3}{2}\left(e_b\chi^{(3),\,\mathrm{int}} : e_b e_a e_b\right) a_a^2 L\,. \tag{20}$$

Comparing (19) and (20) we derive that $\chi_{\mathrm{eff}}^{(3),\,\mathrm{casc}} = \frac{8}{3n_c}\left(d_{\mathrm{eff}}\right)^2 \frac{L}{\lambda_c}$. The longer is the nonlinear media the bigger is the effective cubic nonlinearity.

We would like to point out that the used terminology "cascade" is somehow misleading. The word "cascade" should be used if the second order processes occur one after another as it is in the case of third harmonic generation in two crystals in a row. Here the second order processes take place simultaneously and cannot be separated.

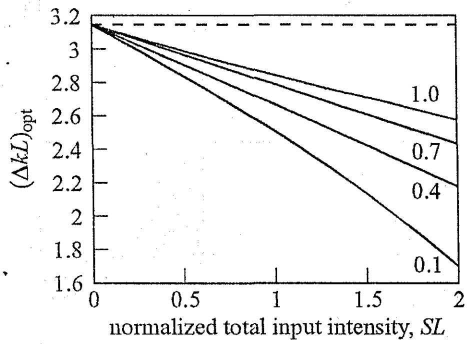


Fig. 3. Optimum values for the phase mismatch ΔkL versus the "normalized total input intensity" $SL^2 = \sigma_c L^2\left[\sigma_b a_a^2(0) + \sigma_a a_b^2(0)\right]$ for different values of the ratio $\frac{\sigma_a a_b^2(0)}{\sigma_b a_a^2(0)}$. Dashed line is obtained from fixed intensity approximation (expression (18)). Solid line is the numerical solution of system (8–10)

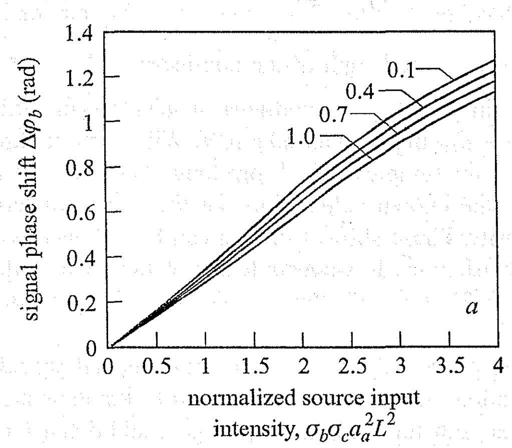
In Fig. 4 is shown the numerical solution for the dependences of the phase shift $\Delta\varphi_b$ (*a*) and the depletion (*b*) of the signal as a function of the input intensity of the wave a for constant ΔkL equal to 2.8. It is seen that $\Delta\varphi_b$ depends almost linearly on the intensity of the wave "a". The smaller is the ratio between the intensities of the signal and the source waves the bigger is the phase shift gained by the signal wave. Nevertheless one order of magnitude change of the input signal results only in 20 % change of the phase shift for given input source intensity.

Phase modulation of the waves gained in nonlinear optical interactions is usually described in terms of nonlinear index of refraction $\bar{n}_2$. In the case of sum frequency mixing

$$n_b = n_{b0} + \Delta n^{\mathrm{int}} + \Delta n^{\mathrm{casc}} = n_{b0} + \Delta n^{\mathrm{int}} + \bar{n}_2^{\mathrm{casc}} I_a\,. \tag{21}$$

The first nonlinear term is a result of direct cubic processes $\omega_b = \omega_b + \omega_b - \omega_b$, $\omega_b = \omega_a + \omega_b - \omega_a$ and $\omega_b = \omega_c + \omega_b - \omega_c$ described by the pure cubic nonlinearity

$\chi^{(3),\,\mathrm{int}}$. The estimations showed that for values of ΔkL close to the optimum, this term can be neglected in comparison to the cascade one.



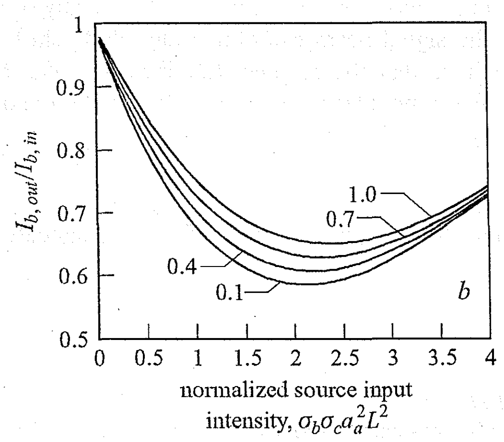


Fig. 4. Numerical results for the induced phase shift (a) and the depletion (b) of the signal wave "b" as a function of the "normalized input intensity" of the source wave "a" for different values of the "normalized input intensity" $\sigma_a a_b^2(0) L^2$ of the signal wave "b"

This last (cascade) term is arising from the simultaneous action of coupled second order processes in the nonlinear media $\omega_c = \omega_a + \omega_b$ and $\omega_b = \omega_c - \omega_a$ and is responsible for the predicted large cross phase modulation $\Delta\varphi_b = \dfrac{2\pi}{\lambda_b}\Delta n^{\mathrm{casc}} L$.

Using (18) we find

$$\bar{n}_2^{\mathrm{casc}} = \frac{4\pi\,(d_{\mathrm{eff}})^2}{\lambda_c n_a n_b n_c \varepsilon_0 c}\frac{\Delta k}{K^2}\left[1 - \mathrm{sinc}(KL)\right] . \tag{22}$$

This expression for small values of S ($\frac{2\sqrt{S}}{\Delta k} \ll 1$) coincides with the expression for $\bar{n}_2^{\text{casc}}$ responsible for self phase modulation found in [7, 9]. The maximal nonlinear index of refraction is proportional to the length of the nonlinear media: $\bar{n}_2^{\text{casc}} = \frac{4\,(d_{\text{eff}})^2}{n_a n_b n_c \varepsilon_0 c}\frac{L}{\lambda_c}$.

As it is pointed out in [18] the new nonlinear noncentrosymmetric organic materials can be designed to have d_{eff} bigger than 50 pm/V. With such nonlinear materials $\bar{n}_2^{casc}$ is about 10^{-11} cm^2/W for samples only 1 mm long. This value is two-three orders of magnitude larger than the known value of $\bar{n}_2$ for the optical materials with electronic origin of the nonlinearity. Phase shift of 0.5 rad can be obtained with 1 ps long pulses with energy density 80 nJ/mm^2. In waveguide applications where the beam is confined to an area of about 10–30 μm^2 only few picojoules will be required for such phase shifts.

Our analysis of the phase behaviour of the fundamental waves in the process of sum frequency generation shows that as a result of simultaneous action of coupled second order processes both fundamental waves gain additional large phase shift. The phase shift of any of the fundamental waves can be controlled by the intensity of the second fundamental wave. In comparison with the scheme using type I second harmonic generation, where for high phase shift is required high intensity of the signal beam with the proposed scheme the signal beam can obtain any phase shift independently on its input intensity. We believe that the scheme described here can be used for efficient ultrafast opto-optical phase modulation, amplitude modulation and deflection.

Acknowledgments

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