Cascaded nonlinear optical processes

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I. Introduction

Cascaded nonlinear optical processes (CNOP) play important role in last decade developments of nonlinear optics. Many of the experiments for generation of extreme ultraviolet and X ray radiation are accompanied by generation of low number harmonics that is sign of involvement of CNOP (Misoguti *et al*. 2001). The last experimental achievements in the attempt to form stable 2D and 3D solitary waves in quadratic crystals are based on the great progress in the understanding of the cascade processes (Stegeman and Torner 2001). The devices based on frequency shifting in quadratic crystals have reached commercial application in optical communication industry (Chou *et al*. 1998, Gallo and Assanto 1999) and are an excellent example of the importance of CNOP.

Cascading in nonlinear optics is an effect noticed during the first steps of nonlinear optics ((Akhmanov and Khokhlov 1964, Ostrovskii 1967), but only recently it received the attention adequate to its importance. In fact CNOP are accompanying effects for all known nonlinear optical processes. As an example, the well known effect - the depletion of the fundamental wave during the process of phase-matched second harmonic generation is cascaded process. Indeed, depletion of fundamental wave is based on two steps: (i) generation of second harmonic wave and (ii) reconstruction of the fundamental wave by difference frequency mixing. Another very simple experiment demonstrating cascaded processes can be done with a quadratic crystal tuned to phase-matched second harmonic generation. If we block fundamental and the SH wave after the crystal and allow the shorter wavelengths to go through we can record not so small third harmonic signal and also forth and fifth harmonic with the decreasing efficiencies. This generation of high-order harmonics is a result of CNOP.

The cascaded nonlinear optical processes can be separated in two big groups. In the first one, due to simultaneous action of two or more sub-processes, the fundamental (input) waves change their parameters like amplitude, phase, polarization state, beam size, temporal shape. The second group includes these cascaded interactions in which new waves are generated. In its biggest part the cascaded interactions simulate direct $\chi^{(3)}$ or direct $\chi^{(5)}$ processes. The advantage of CNOP is its higher efficiency and extremely short time constant. The cascading effects can be used for construction of nonlinear optical devices for efficient high order harmonics generation, for construction of devices for optical communications, all optical switching devices, mode locking devices and etc.

The other possible classification of the nonlinear optical cascaded processes is based on the number of sub-processes involved in the cascading. There are two possibilities: all sub-processes are governed by one and the same mismatch parameter and the case when several mismatch parameters control the whole cascaded process. An example for cascaded process that belongs to the first type is the cascading connected with the process of second harmonic generation. The efficiencies of the two cascading steps $\omega + \omega = 2\omega$ and $2\omega - \omega = \omega$ depend on *single* phase-matching parameter

$\Delta k_2 = k_2 - 2k_1$ (k_1, k_2 are the wave vectors of the fundamental and second harmonic wave, respectively). The effect of third harmonic generation in single quadratic crystal, where the two cascading steps are second harmonic generation and sum frequency mixing, is an example of the cascaded process that is controlled by *two* phase-matching parameters: $\Delta k_2 = k_2 - 2k_1$ and $\Delta k_3 = k_3 - k_2 - k_1$ (k_3 is the wave vector of the third harmonic wave).

Above we gave examples of cascading of second order processes. Indeed, as shown below, cascading of third order effects is also important field of nonlinear optics of cascaded processes. In a similar way, third order cascading can be observed inside single third order interaction or be a result of cascading of different third order processes. We will consider in this chapter third order cascading as well.

II. Cascading controlled by single phase mismatched parameter

II.1 Cascading with Type I SHG

Let us consider the cascading effects connected with the most popular nonlinear interaction – second harmonic generation – Type I. The cascading steps are two: $\omega + \omega = 2\omega$ and $2\omega - \omega = \omega$. In both steps second harmonic generation (SHG) and the difference frequency mixing (DFM) are involved only two waves fundamental and second harmonic (SH) wave. So, both sub-processes depend only on one phase-mismatch parameter $\Delta k_2 = k_2 - 2k_1$. We will show that due to cascading the fundamental wave accumulate during the interaction nonlinear phase shift (NPS) – the effect that in spite of some early works was indeed realized last decade (Stegeman *et al.* 1996)

The plane wave equations describing the process of SHG are:

$$\frac{dA_1}{dz} = -i\sigma A_2 A_1^* \exp(-i\Delta k_2 z), \tag{1a}$$

$$\frac{dA_2}{dz} = -i\sigma A_1^2 \exp(i\Delta k_2 z) . \tag{1b}$$

Representing the complex amplitudes A_1 and A_2 in the form

$$A_1 = a_1 \exp(i\varphi_1),$$

$$A_2 = a_2 \exp(i\varphi_2)$$

and introducing the phase

$$\Phi = \varphi_2 - 2\varphi_1 - \Delta k_2 z$$

we can express the phase of the fundamental wave and the module of the SH wave by following differential equations

$$\frac{d\varphi_1}{dz} = -\sigma a_2 \cos\Phi , \tag{2a}$$

$$\frac{da_2}{dz} = -\sigma a_1^2 \sin\Phi . \tag{2b}$$

With the help of two invariants of the system

$$\sigma a_1^2 a_2 \cos\Phi + \frac{\Delta k_2}{2} a_2^2 = \Gamma = 0 \quad , \tag{3a}$$

$$a_1^2 + a_2^2 = a_{10}^2 + a_{20}^2 = u^2 \tag{3b}$$

we obtain

$$\frac{da_2^2}{dz} = -2\sigma u^3 \sqrt{\left(\frac{a_2}{u}\right)^2 - \frac{1}{M}\left(\frac{a_2}{u}\right)^4 + \left(\frac{a_2}{u}\right)^6} \; . \tag{4}$$

Neglecting last term in the square root in (4) and after integration of (4) we get the expressions for the efficiency of the SH and fundamental waves:

$$\left(\frac{a_1}{u}\right)^2 = 1 - M\sin^2(Qz)\,, \tag{5a}$$

$$\left(\frac{a_2}{u}\right)^2 = M\sin^2(Qz)\,, \tag{5b}$$

where

$$M = (\sigma u/Q)^2 \;,\; Q = \sqrt{2\sigma^2 u^2 + \Delta k_2{}^2/4} \; .$$

With known a_1 and a_2 Equation 2a can be integrated that results in

$$\Delta\varphi_{NPS} = \frac{\Delta k_2 L}{2}\left(\frac{\arctan\left[\sqrt{1-M}\tan(QL)\right]}{QL\sqrt{1-M}} - 1\right) . \tag{6}$$

Expresions (5) and (6) describe the cascading effects for not very big conversion efficiencies (<30%): the depletion of the fundametal wave and the accumulation of the NPS by the fundamental wave. These dependencies are shown on Figure 1a,b.

For bigger input fundamental intensities the adequate description of the phase and the amplitudes require numerical solution of the system (1). We see from the shown curves that at phase-mismatches $\Delta k_2 L$ "+" or "-" π the NPS accumulated by the fundamental wave has maximum. The remarkable point is that the sign of the NPS can be changed by the simple change of sign of the phase-mismatch parameter. In the real experiments the change or Δk_2 can be done by changing e. g. the angular position of the crystal or change of the temperature. The higher is the input fundamental intensity, the bigger is the accumulated NPS.

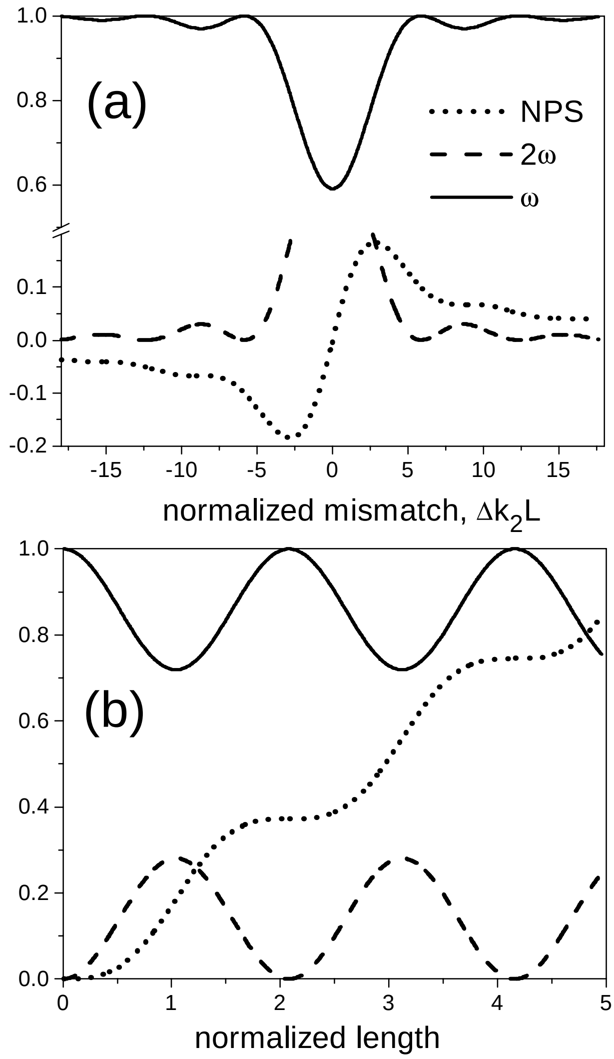

Figure 4. *a) Fundamental depletion , second harmonic efficiency and nonlinear phase shift versus normalized mismatch* $\Delta k_2 L$*,* $[\sigma u L = 0.8]$ *and b) the same parameters versus normalized length of the crystal* $\sigma u L$*,*$[\Delta k_2/\sigma u = 2.5]$ *.*

In order to demonstrate that this type of cascading is in fact simulation of third order effect and can be described in terms of $\chi^{(3)}_{casc}$ let us present (6) in different form. Expressing $\cos\Phi$ from (3a) and substituting it in (2a) and using the amplitude of SH wave defined in (5b) after integration of (2) we obtain :

$$\Delta\varphi_{NPS} = \Delta k_2 L\left(\frac{\sigma u}{2Q}\right)^2\left[1-\frac{\sin(2QL)}{2QL}\right],$$

that for really small input intensities $(\sigma uL << 1)$ has the form

$$\Delta\varphi_{NPS} = \left(\frac{\sigma u}{\Delta k}\right)^2\left[\Delta k_2 L - \sin(\Delta k_2 L)\right]. \tag{7}$$

For $\Delta k_2 L = \pi$ and keeping in mind that $\sigma = \frac{2\pi d^{(2)}}{\lambda n} = \frac{\pi\chi^{(2)}}{\lambda n}$ we find that accumulated by the fundamental wave NPS is proportional to the square of the second order susceptibility

$$\Delta\varphi_{NPS} = \frac{\pi u^2 L^2}{\lambda^2 n^2}\left[\chi^{(2)}\chi^{(2)}\right]. \tag{8}$$

The last expression illustrates the origin of the terminology $\chi^{(2)} : \chi^{(2)}$ cascading.

In centrosymmetric media NPS is a result of self-phase modulation due to inherent $\chi^{(3)}$ nonlinearity:

$$\Delta\varphi_{NPS} = \frac{3\pi u^2 L}{4\lambda\, n}\chi^{(3)}. \tag{9}$$

Comparing Equations (8) and (9) we find

$$\chi^{(3)}_{casc} = \frac{4L}{3\lambda n}\left[\chi^{(2)}\chi^{(2)}\right], \tag{10}$$

$$n_{2,casc} = \frac{L}{2\lambda n^2}\left[\chi^{(2)}\chi^{(2)}\right] \tag{11}$$

The main conclusion at this point is that cascading in quadratic media can simulate cubic processes and we can expect many of the known observed on the base of $\chi^{(2)} : \chi^{(2)}$ cascading. Since $\chi^{(2)}$ effects are instantaneous, not connected with the real transitions in the media then the $\chi^{(3)}_{casc}$ is also instantaneous.

II.2 Cascading with Type II SHG

With this type of SHG we have two input waves: ordinary and extraordinary. When the goal is maximum SH efficiency it is usually chosen ordinary and extraordinary wave to have equal intensity.

For obtaining maximum effect from the second order cascading with Type II interactions it is required ordinary and extraordinary wave to have *unequal* intensity. The plane waves equations describing Type II SHG are:

$$\frac{dA_{1o}}{dz} = -i\sigma A_2 A_{1e}^{*} \exp\ (-i\Delta kz)\ ,$$

$$\frac{dA_{1e}}{dz} = -i\sigma A_2 A_{1o}^{*} \exp(-i\Delta kz)\ , \qquad (12)$$

$$\frac{dA_2}{dz} = -i2\sigma A_{1e} A_{1o} \exp(i\Delta kz)\ ,$$

The involved waves in this quadratic interaction are three: ordinary and extraordinary fundamental waves and SH wave. Respectively, the cascading steps are three:

$$\omega_o + \omega_e = \omega_{2e}; \omega_{2e} - \omega_o = \omega_e;$$

$$\omega_{2e} - \omega_e = \omega_o.$$

Single phase-matching parameter $\Delta k = k_{2e} - k_o - k_e$ is controlling all cascading steps.

Similar way, as in II.1, we derive appoximate "low intensity" expressions for the NPS collected by both fundamental waves (Saltiel *et al* 1995, 1996):

$$\Delta\varphi_{1o} = \left(\frac{\sigma u_{1e}}{\Delta k}\right)^2 [\Delta kL - \sin(\Delta kL)],$$

$$\Delta\varphi_{1e} = \left(\frac{\sigma u_{1o}}{\Delta k}\right)^2 [\Delta kL - \sin(\Delta kL)] \qquad (13)$$

It is important to note that the intensity of the ordianry wave at the input of the crystal defines the NPS of the extraordinary wave and vice versa.

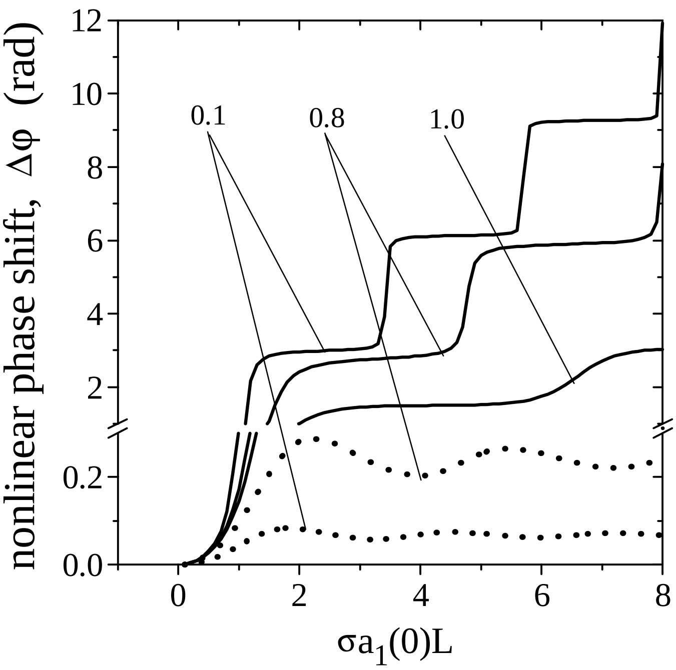

Figure 2. *Nonlinear phase shift collected by the less intensive (solid lines) and more intensive (doted lines) fundamental waves involved in the process of Type II SHG as a function of normalized input amplitude of the more intensive fundamental wave. The parameter is the ratio of the intensities of the two fundamental waves. The phase-matching parameter is* $\Delta kL = 0.3$.

Figure 2 (obtained by numerical integration of System (12)) shows nonlinear phase shift collected by both fundamental waves for ratio 10 of their input intensities. The less intensive wave accumulates the bigger NPS. This effect is used for construction of all-optical transistors (Assanto *et al.* 1995) and for many polarization devices as described below.

II.3. Evolution of the polarization state as a result of $\chi^{(2)} : \chi^{(2)}$ cascading in Type II SHG.

As an interesting example of the capability of second order cascading to simulate third order processes we consider change of the polarization state of wave involved in Type II SHG (Buchvarov *et al.* 1997). Simulated cubic effect is induced polarization rotation and induced ellepticity observed along four-fold axis in cubic centrosymmetric crystals. In this cubic effect change of the polarization effect is a result of anisotropy of the third order susceptibility. We show here that the same effect can be observed in quadratic crystals due to cascaded effects. The idea of effect is shown on Figure 3. Change of the polarization state of the input fundamental wave involved in Type II SHG is a result of the effect of the asymmetry of the accumulation of NPS with respect to the ordinary and extraordinary waves considered in the previous paragraph. Let us first note that linear birefringence has to be compensated by proper wave plate (not shown). Then, if the NPS collected by the two waves is different – this will lead to change of the polarization state at the output of the quadratic crystal. We have to remember also that the depletion of the two eigen waves (ordinary and extraordinary) will also influence the output polarization state.

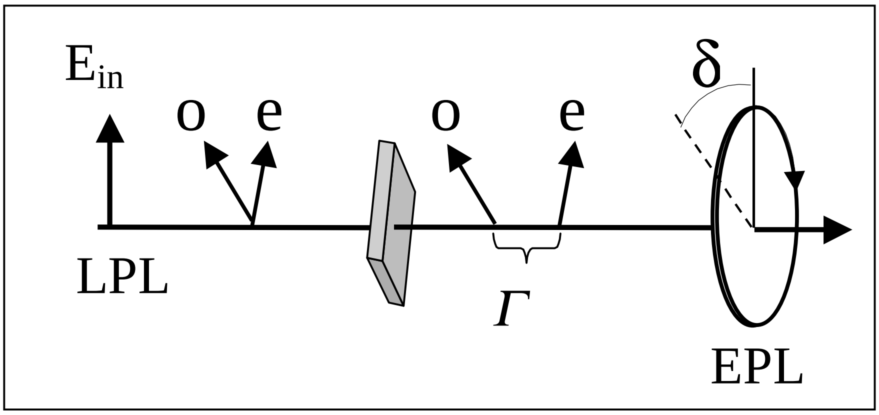

Figure 3. *Induced change of the polarization state as a result of second order cascading in Type II SHG crystal. LPL – linearly polarized light; EPL – elliptically polarized light; Γ – accumulated nonlinear phase shift difference between ordinary and extraordinary wave.*

The output complex amplitudes of the two fundamental waves as a result of cascading became

$$E_{out}^{(o)} = E_{in}^{(o)}\rho_o \exp\left(i\Delta\varphi_{NPS}^{(o)}\right)$$
$$E_{out}^{(e)} = E_{in}^{(e)}\rho_e \exp\left(i\Delta\varphi_{NPS}^{(e)}\right), \qquad (14)$$

where ρ_o and ρ_e are the amplitude transmission coefficients of the two eigen waves.

If we introduce the complex ratio $R = \dfrac{E_{out}^{(o)}}{E_{out}^{(e)}}$ then the induced ellipticity ε and induced azimuth rotation δ_o can be calculated using following relations (Azzam and Bashara 1977)

$$\tan(2\delta_o) = 2\,\mathrm{Re}(R)\Big/\left(1-|R|^2\right) = \frac{\rho_o\rho_e\cos(\Gamma)\sin(2\alpha)}{\rho_e^2\sin^2(\alpha)-\rho_o^2\cos^2(\alpha)} \qquad (15a)$$

$$\sin(2\varepsilon) = 2\,\mathrm{Im}(R)\Big/\left(1+|R|^2\right) = \frac{\rho_o\rho_e\sin(\Gamma)\sin(2\alpha)}{\rho_e^2\sin^2(\alpha)+\rho_o^2\cos^2(\alpha)} \qquad (15b)$$

In Equations (15ab) α is the angle between input polarization direction and the polarization plane of the ordinary wave and $\Gamma = \Delta\varphi_{NPS}^{(o)} - \Delta\varphi_{NPS}^{(e)}$.

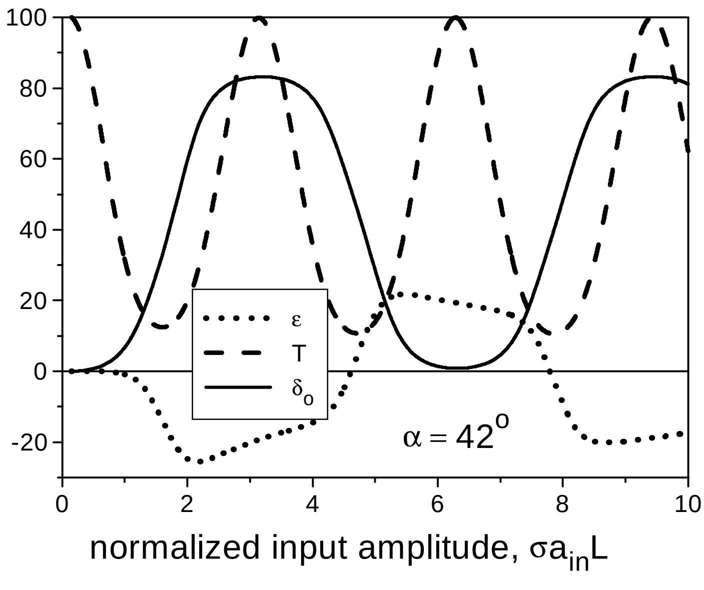

Figure 4. *The angle of ellipticity ε, the rotation angle δ_o and the transmission of the fundamental wave T (in %) as a function of the normalized input amplitude for the indicated input angle α. The normalized phase-mismatch $\Delta kL = 0.3$.*

On Figures 4 are shown these two parameters and the transmission of the fundamental wave as a function of the input amplitude. The important factor for observation of these induced effects is the input amplitudes for the ordinary and extraordinary waves not to be equal ($\alpha \neq 45^o$).

Similar effect can be observed also with Type I SHG. If on the crystal is falling linearly polarized wave the crystal should be rotated around laser beam axis such a way so input wave is splitted into ordinary and extraordinary wave with equal intensity. Since only ordinary wave accumulates NPS and experience depletion and the ordinary wave is not involved in the process of SHG if the interaction is $o_1 o_1$-e_2 . then $\rho_e = 1$ and $\Delta\varphi_{NPS}^{(e)} = 0$. Then we can use again (14) and (15) to calculate the induced change the polarazation state.

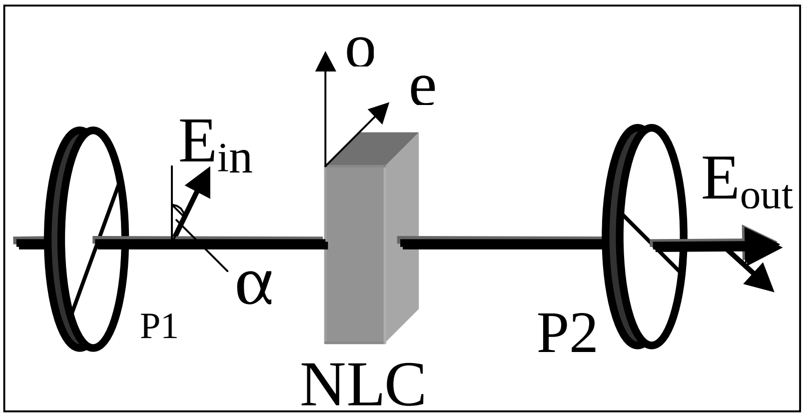

Figure 5. *Frequency doubling polarization interferometer. NLC – nonlinear crystal for SHG; P1 – polarizer; P2 - analyzer . α – the angle between input polarization and polarization plane of the ordinary wave*

II.4. Polarization interferometer based on $\chi^{(2)} : \chi^{(2)}$ cascading.

The effect of intensity induced polarization rotation can be used for construction of devices in which the frequency doubling crystal(s) is sandwiched between two polarizers. The scheme of such a device, some times called frequency doubling polarization interferometer (Saltiel *et al.* 1996b, Buchvarov *et al.* 1997) is shown on fig. 5. Both Type I and Type II frequency doubling schemes can be used; the device consists also phase corrector (not shown). The angle α is the angle between the input polarization plane and the direction of the ordinary wave polarization vector. Depending on the suggested application the polarization planes of the two polarizers can be perpendicular, parallel or construct an angle 2α. The phase corrector is used to compensate the linear phase shift that the two waves obtain in the crystal. Depending on the mutual orientation of the polarizer, analyzer and the frequency doubling crystal, the nonlinear polarization interferometer exhibits self induced transparency or self induced darkening. The throughput of the frequency doubling polarization interferometer (FDPI) for the three different arrangement of the FDPI is given by

$$\begin{aligned}
T_{\perp} &= \frac{1}{4}\left(\rho_o^2 + \rho_e^2 - 2\rho_o\rho_e \cos\Gamma\right)\sin^2(2\alpha),\\
T_{II} &= \rho_o^2 \cos^4\alpha + \rho_e^2 \sin^4\alpha + \frac{1}{2}\rho_o\rho_e \cos\Gamma \sin^2(2\alpha),\\
T_{2\alpha} &= \rho_o^2 \cos^4\alpha + \rho_e^2 \sin^4\alpha - \frac{1}{2}\rho_o\rho_e \cos\Gamma \sin^2(2\alpha),
\end{aligned} \tag{16}$$

where as before $\Gamma = \Delta\varphi_{NPS}^{(e)} - \Delta\varphi_{NPS}^{(o)}$ and ρ_o and ρ_e are the amplitude transmission coefficients for the ordinary and extraordinary wave, respectively

The third expression describe the "2α" version of the interferometer - the situation when analyzer construct angle 2α with the polarizer .The transmission curves of three different versions of

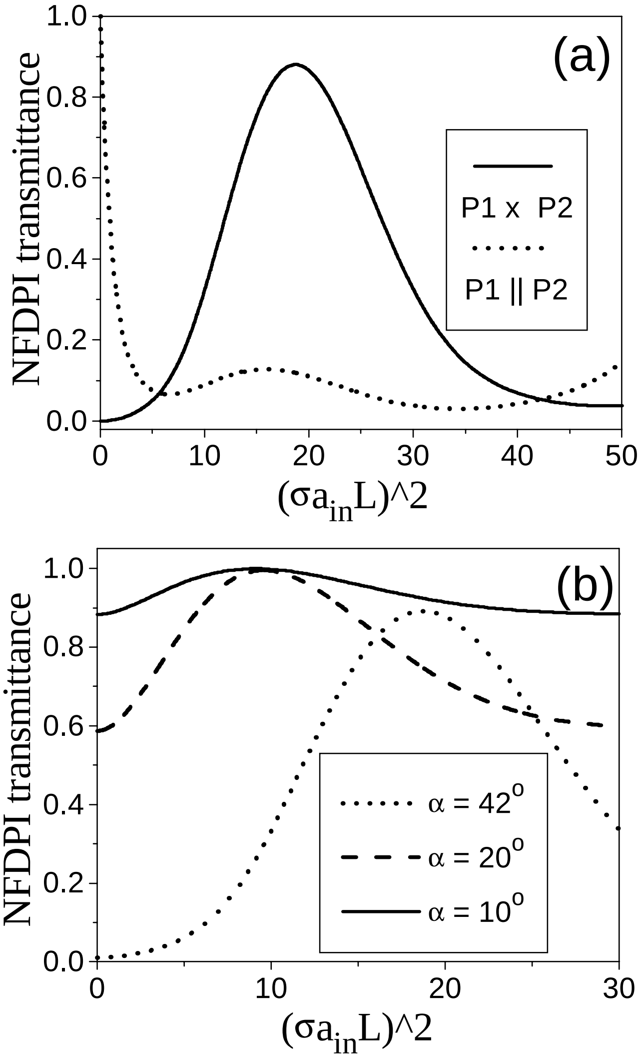

Figure 6. *Transmittance of the FDPI as a function of normalized input intensity for tree different arrengments: a) for parallel and crossed polarizers and b) for polarizers that constitute angle* 2α *.The phase-mismatch is* $\Delta kL = 0.3$.

the FDPI, shown on Figure 5 is presented on Figure 6. When the polarizer and the analyzer are crossed, strong shortening of the pulses can be obtained (Buchvarov *et al.* 1997).

It is clear that the two capabilities of the FDPI: pulse shortening and self induced transparency effect at relatively low power (we calculate 60 MW/cm^2 for 10 mm long KTP crystal) make this device suitable for mode locking of lasers. Another possible application could be all optical switching, and sensor protection.

Experimentally FDPI has been realized with bulk phase matched KTP (Lefort and Barthelemy 1995, Louis *et al.* 2001). As seen from Figure 6b by varying the angle α between 0 and $\pi/4$ with "2α" version of the interferometer one can achieve any contrast ratio is between open and closed working points. Flexibility to change the contrast ratios very important when this effect is used for mode-locking of lasers. Successful mode-locking with such an interferometer is reported by (Louis *et al.* 2001)

II.5 Cascading in centrosymmetric media

As we demonstrate in the previous paragraphs $\chi^{(2)}:\chi^{(2)}$cascading simulates cubic effects and causes self-interaction effects known as "n_2" effects typical for cubic interactions, like accumulation of nonlinear phase shift, self-focusing and self defocusing, change of polarization stage, solitons and etc. The question is – is it possible cascading effects to be observed in centrosymmetric media as a result of cascading of third order processes. Let us consider centrosymmetric media with the possibility for third harmonic generation $\omega+\omega+\omega=3\omega$ with very small phase-mismatched parameter $\Delta k = k_3 - 3k_1$. Generated third harmonic wave interacts again with the fundamental wave through four wave mixing down conversion process $3\omega-\omega-\omega=\omega$. Reconstructed through this two step cascading fundamental wave is phase shifted with amount that depends on amount of the phase-mismatch parameter Δk. Such a way fundamental wave at the output experiences NPS. The resulted phase shift depends on the square of input intensity indicating that this cascading effect has "n_4" behavior simulating artificially the role of $\chi^{(5)}$in the process of NPS collection in centrosymmetric media.

The plane wave equations describing the effect of third harmonic generation are:

$$\frac{dA}{dz} = -i\left(\gamma_1|A|^2 + \gamma_2|T|^2 + \delta|A|^4\right)A - i\gamma_{13}T\left(A^*\right)^2 \exp(-i\Delta kz) \tag{17a}$$

$$\frac{dT}{dz} = -i\left(\gamma_4|A|^2 + \gamma_5|T|^2\right)T - i\gamma_{31}A^3 \exp(i\Delta kz) \tag{17b}$$

where $\gamma_{13} = \frac{9\omega}{8c}\frac{\chi^{(3)}}{n_1}$ and $\gamma_{31} = \frac{9\omega}{8c}\frac{\chi^{(3)}}{n_3}$ and $\gamma_1, \gamma_2, \gamma_3, \gamma_4$ and δ are nonlinear coupling coefficients responsible for self- and cross-phase modulation.

To obtain the effect of third order cascading, however, one has to solve them (applying the same procedure as in II.1) with respect to the phase of the fundamental beam (Saltiel *at al.* 1997). In approximation of not big conversion efficiency into third harmonic generation the output phase of the fundamental wave is

$$\begin{aligned} \Delta\varphi_1^{NPS} &= \Delta\varphi_{1,dir}^{NPS} + \Delta\varphi_{1,casc}^{NPS} \\ \Delta\varphi_{1,dir}^{NPS} &= \left[\gamma_1 a_{10}^2 + \delta a_{10}^4\right]L \end{aligned} \tag{18}$$

with

$$\Delta\varphi_{1,casc}^{NPS} \approx -\gamma_{13}\gamma_{31}\frac{a_{10}^4}{(\Delta k)^2}\left[\Delta kL - \sin(\Delta kL)\right] \tag{19}$$

Two important conclusions we can deduce from the result for the NPS given by (19). The NPS has quadratic dependence on the input intensity $I_1 \propto a_{10}^2$. The product $\gamma_{13}\gamma_{31} \propto \chi^{(3)}.\chi^{(3)}$ directly indicates that the NPS is result of third order cascading, called also $\chi^{(3)} : \chi^{(3)}$ cascading. As in second order cascading the sign of the NPS due to third order cascading depends on the sign of phase-mismatch parameter Δk. By comparing with formula (7) we see the dependence of the nonlinear phase shift on the phase mismatch parameter is the same as for $\chi^{(2)} : \chi^{(2)}$ cascading . In fact the expression (19) can be considered as an additional term $n_4^{casc}E^4$ in the expansion of the index of refraction on the power of the electric field.

$$n = n_o + n_2E^2 + n_4E^4 + n_4^{casc}E^4 + \ldots \tag{20}$$

where n_4^{casc} is defined as

$$n_4^{casc}(\max) \approx -sign(\Delta k).2n_2^2\frac{L}{\lambda} \quad . \tag{21}$$

This type new kind saturating nonlinearities – non- Kerr nonlinearities – arising from phase-matched interaction between the fundamental beam and its third harmonic can have a dramatic effect on the propagation of spatial solitary waves. Conditions for propagation of solitary waves (both bright and dark) in centrosymmetric media in presence of third harmonic wave are found (Kivshar 1998).

III. Cascading controlled by several phase-mismatching parameters.

In this part we consider the second type nonlinear cascaded processes that involve several processes, each of them with its own phase mismatched parameter. Here we show that these processes play important role in high order harmonic generation, frequency shifting processes, nonlinear light scattering, generation of cross polarized waves and nonlinear polarization rotation. Cascaded interactions controlled by several phase-mismatching parameters have been observed in both centrosymmetric and non-centrosymmetric media.

In quadratic media the most efficient are cascaded interactions that start with SHG process. Indeed recently big interest attracted the single crystal cascaded interactions listed in the table below:

Cascaded interaction	First $\chi^{(2)}$ step	Second $\chi^{(2)}$ step	Equivalent process
Third harmonic generation	$\omega+\omega=2\omega$	$\omega+2\omega=3\omega$	$\omega+\omega+\omega=3\omega$
Fourth harmonic generation	$\omega+\omega=2\omega$	$2\omega+2\omega=4\omega$	$\omega+\omega+\omega+\omega=4\omega$
Generation of cross polarized wave	$\omega+\omega=2\omega$	$2\omega-\omega=\omega_\perp$	$\omega+\omega-\omega=\omega_\perp$
Frequency shifting (frequency conversion)	$\omega+\omega=2\omega$	$2\omega-\omega_a=\omega_b$	$\omega+\omega-\omega_a=\omega_b$

Table 1 . *Two step cascaded nonlinear optical interactions starting with second harmonic generation.*

Third harmonic generation in single quadratic crystal is the most studied cascaded interaction controlled by two phase-mismatching parameters and we start with this interaction.

III.1 Third harmonic generation in single quadratic crystal

Third harmonic generation (THG) in single non centrosymmetric crystal can be result of direct process $\omega+\omega+\omega=3\omega$, or result of cascading of two second order processes $\omega+\omega=2\omega$ and $2\omega+\omega=3\omega$. When the quadratic processes are close to their exact phase matching the direct process is relatively weak and can be neglected and the efficiency of the generated THG is result mostly of the second order cascading. THG in single $\chi^{(2)}$ crystal can occur under four different phase matching conditions. They can be easily found from plane wave equations describing this process:

$$\frac{dA_1}{dz}=-i\sigma_1 A_2 A_1^*\exp(-i\Delta k_{SHG}z)-i\sigma_2 A_3 A_2^*\exp(-i\Delta k_{SFM}z) \quad ,$$

$$\frac{dA_2}{dz}=-i\sigma_1 A_1^2\exp(i\Delta k_{SHG}z)-i2\sigma_2 A_3 A_1^*\exp(-i\Delta k_{SFM}z) \quad , \tag{22}$$

$$\frac{dA_3}{dz} = -i3\sigma_2 A_2 A_1 \exp(i\Delta k_{SFM} z) \quad ,$$

where $\Delta k_{SFG} = k_3 - k_2 - k_1 \quad \Delta k_{SHG} = k_2 - 2k_1$.

Indeed if we neglect the depletion of the fundamental and the second harmonic wave the system (22) simplifies to system (23) that can be easily solved analytically

$$\frac{dA_1}{dz} = 0$$
$$\frac{dA_2}{dz} = -i\sigma_1 A_1^2 \exp(i\Delta k_{SHG} z) \qquad (23)$$
$$\frac{dA_3}{dz} = -i3\sigma_2 A_1 A_2 \exp(i\Delta k_{SFG} z).$$

The four conditions that allow generation of efficient third harmonic generation are as follow:

i) when the phase matching for SHG is fulfilled :

$$\Delta k_{SHG} = k_2 - 2k_1 \to 0.$$

The amplitude of the third harmonic wave is

$$A_3(z) = \frac{i3\sigma_1\sigma_2 A_1^3 z}{\Delta k_{SFG}} \exp(i\Delta k_{SFG} z); \quad (24)$$

ii) when the phase matching for SFM is fulfilled:

$$\Delta k_{SFM} = k_3 - k_2 - k_1 \to 0.$$

The amplitude of the third harmonic wave is

$$A_3(z) = -\frac{i3\sigma_1\sigma_2 A_1^3 z}{\Delta k_{SHG}} \quad ; \qquad (25)$$

iii) when the phase matching for direct THG process $\omega + \omega + \omega = 3\omega$ is fulfilled:

$$\Delta k_{SFM} + \Delta k_{SHG} = k_3 - 3k_1 \to 0.$$

This is the only condition when the amplitude of the TH wave will be result of interference of the cascaded contribution and direct contribution based on inherent $\chi^{(3)}$ nonlinearity $(\gamma \propto \chi^{(3)})$ in the media .

$$A_3(z) = i\left(\frac{3\sigma_1\sigma_2}{\Delta k_{SHG}} + \gamma\right) A_1^3 z \qquad (26)$$

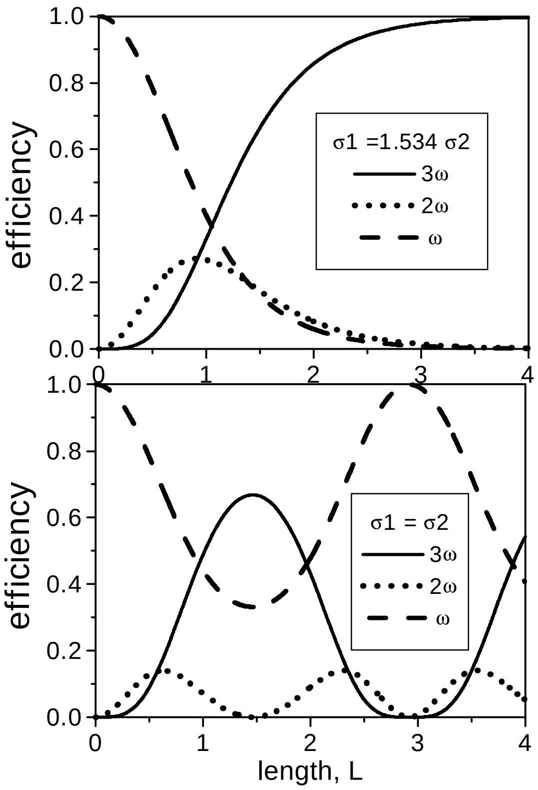

Figure 7. *Calculated by numerical integration of system (22) efficiency of third and second harmonic waves and the depletion of the fundamental wave as a function of normalized length of the crystal* $\sigma_1 a_{1o} L$ *for the case of exact phase matching for the two cascaded steps and different ratios for* σ_1/σ_2 *as indicated.*

For the three cases considered above the generated TH intensity depends on the square of length of the nonlinear media and on the cub of the input intensity. In the literature the biggest efficiency reported is 6% measured in single BBO crystal for the (iii) case ((Banks *et al.* 1999)).

The most efficient and interesting case is when both quadratic steps are simultaneously phase matched

(iv) $\Delta k_{SFM} = k_3 - k_2 - k_1 \to 0$ and $\Delta k_{SHG} = k_2 - 2k_1 \to 0$

then the intensity of the third harmonic wave will depend on the four power of the length of the nonlinear media!

$$\left|A_3(z)\right|^2 = (3\sigma_1\sigma_2)^2 \left|A_1\right|^6 z^4 \qquad (27)$$

For realization of this fourth case (iv) it is required achievements of simultaneous phase matching of two processes. The methods for do that will be discussed below in chapter IV. The highest reported efficiency of THG in condition of double phase matching in LiTaO3 is as high as 27% ((Zhang *et al* 2001).

For higher conversion efficiencies is no longer correct to neglect the depletion of the fundamental and second harmonic wave. In this case direct numerical integration of (22) is required. In general the solution gives periodical change of the third-harmonic efficiency with the increase the length of the media or input intensity. However for proper chosen ratio of the nonlinear coupling coefficients σ_1 and σ_2 the theoreticaly predicted efficiency is 100% (Egorov and Sukhorukov 1998) as illustrated in Figure 7.

The effect of high order harmonic generation (and in particular THG) in single quadratic crystal is promising for construction of compact frequency converters for diode-pumped solid-state lasers. The second application of these processes is the for measurements of the magnitude cubic nonlinearities. Indeed if the tensors of quadratic nonlinearities are known and we compare the intensity at phase matching conditions (i), (ii) and (iii) we can express the cubic coupling coefficient in terms of second-order coupling coefficients. Cascaded effects allow to build callibration link between high order nonlinearities and low order nonlinearities . Such a way first estimations of $\chi^{(5)}$ in calcite (Akhmanov *et al*. 1975) and $\chi^{(4)}$ in Lithium formiate crystal (Akhmanov *et al.* 1974) have been done. The same way, using different phase matching conditions for single crystal THG, unknown $\chi^{(2)}$ components can be expressed in terms of known $\chi^{(2)}$ components. This was demonstrated in the work of Petrov *et al* (1999), where by measuring polarization anisotropy of cascaded THG in LBO unknown $\chi^{(2)}$ components in this crystal have been measured.

III.2 Cross polarized wave generation in single quadratic crystal.

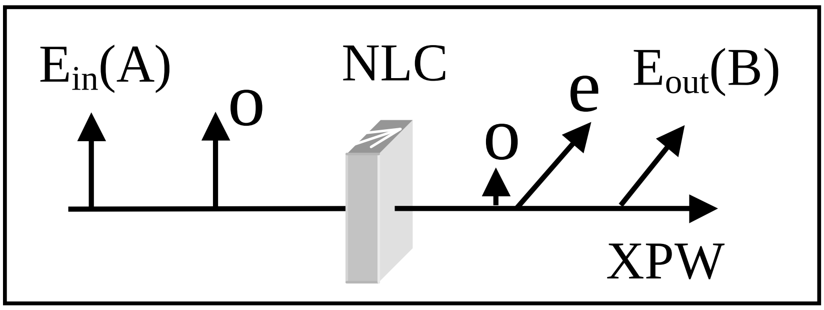

Fig. 8. The idea for cross-polarized wave (XPW) generation via second order cascaded processes. NLC – nonlinear crystal for SHG; o – ordinary; e - extraordinary .

This is another example of second order cascaded interaction controlled by two phase matching parameters listed in Table 1. Indeed (see figure 8), let us consider a quadratic media that supports simultaneously Type I (AA - S) and Type II interaction (AB

– S), where with A and B we mark the fundamental waves with two orthogonal polarizations and S mark the second harmonic field. In the first step with the process AA – S second harmonic wave is generated; with the second step generated second harmonic wave when mixed with the fundamental wave is down-converted by the process SA – B to a wave at fundamental frequency, but with polarization plane perpendicular to the input one. If the both processes are exactly phase matched theoretical prediction is that one can achieve more than 60% conversion from ordinary to extraordinary wave (Saltiel and Deyanova 1999) or vice versa for equal nonlinearities for the two steps. This device can be called "nonlinear porarization rotator". As most of second order cascaded processes the effect of cross-polarized wave (XPW) generation has an equivalent cubic process. With the above notations this is the process AAA* – B. Similar way as for considered cascaded THG in III.1 the process of XPW generation will be efficient for three phase-matched condition: phase-matching for the first step – Type I SHG; phase-matching for the second step – Type II SHG; phase-matching for the direct process $\omega + \omega - \omega = \omega_\perp$. The direct process is automatically phase matched along the z axis of the uniaxial quadratic crystals, where ordinary and extraordinary waves have the same velocities. In this direction however the cascaded process of XPW is interfering with the direct process governed by the inherent $\chi^{(3)}$ nonlinearity of the quadratic crystal.

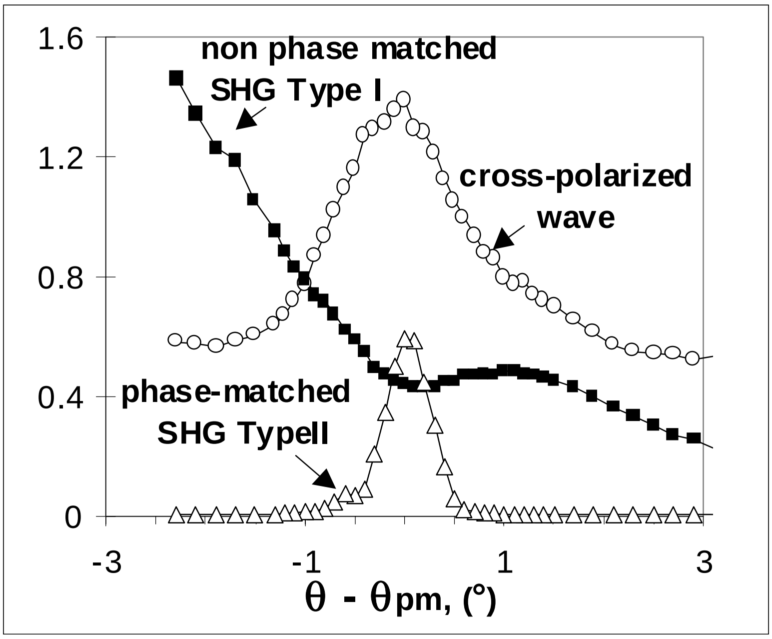

Figure 9. *Experimentally measured cross polarized wave and non-phase matched SH signals (in a.u.) as a function of the deviation of the crystal orientation from the phase matching angle for type II SHG. The bottom curve, represents the phase-matched type II SHG signal measured in a separate experiment. (the purpose of this recording is to mark the position of the phase-matching angle for type II SHG)..*

Experimentaly the process of XPW generation in single quadratic crystal was demonsrated in BBO crystal ((Petrov *et al.* 2001)), one of the crystals that support co-existence of Type I and Type II SHG. One of the processes (AA – S) was not phase matched that is why the measured overall efficiency is small. XPW signal was observed when the crystal was tuned at exact phase matching condition for the second step $\Delta k_{II\ step} = k_{SH} - k_A - k_B \to 0$. The experimental evidence of generated XPW is shown on Figure 9. The fact that the SH wave is depleted exactly at the point of maximum XPW wave efficiency confirms that this "cubic" process is result of two-step second order cascading .

THG ceneration and XPW generation in single quadratic crystal are only two of the all possible cascaded interactions that lead to generation of new waves. Other important interactions are fourth harmonic generation in single crystal (Akhmanov *et al.* 1974, Kildal and Iseler 1978, Hooper *et al.* 1994) and cascaded single crystal frequency conversion that is very efficient analog of the cubic frequency shifting through nondegenerate four wave mixing (Chou *et al*. 1998, Gallo and Assanto 1999).

Cascaded interactions that lead to generation of new waves can be also realized in centrosymmetric media. In this case we have cascading of two third-order processes. Typical example is fifth harmonic generation as a result of cascading of :

a) third harmonic generation $\omega + \omega + \omega = 3\omega$ with phase matching condition $\Delta k_{I\ step} = k_3 - 3k_1$ and

b) four-wave mixing up-conversion $3\omega + \omega + \omega = 5\omega$ with phase matching condition $\Delta k_{II\ step} = k_5 - k_3 - 2k_1$

For the first time such a kind of interaction was realized in $CaCO_3$ by Akhmanov et al (1975). Later many experiments in gases confirmed the important role of cascaded processes in the efforts to generate coherent vacuum ultraviolet and soft X ray radiation by high order harmonic generation (Misoguti *et al*. 2001). In the next paragraph we will consider another type of cascading of two third order processes.

III. 3. Polarization rotation induced by cascading of two third-order processes.

In the II.3 paragraph we showed that in quadratic media CNOP can cause cross-polarized wave generation and change of the polarization state. We find out that the same effect – XPW generation and respectively, induced polarization rotation and change of the polarization state can be caused by cascading of two third order processes in centrosymmetric media when the laser light is propagating along z axis. The two cascading cubic sub-processes are self-phase modulation and four wave mixing.

For describing this effect let us consider, in the slowly varying envelope approximation, plane wave propagation equations, neglecting linear absorption and choosing a coordinate system defined by the directions of the crossed polarizer and analyser. The equations for self-phase modulation of the co-polarized component A and generation of XPW with amplitude B, upon the condition where $|B| << |A|$ (i.e. neglecting the possible effect on cross-phase modulation of wave A caused by the presence of wave B and self-phase modulation of wave B) are (Minkovsli *et al.* 2002):

$$dA/dz = i\gamma_{II}|A|^2 A \quad , \tag{28a}$$

$$dB/dz = i\gamma_{\perp}|A|^2 A \quad , \tag{28b}$$

where $\gamma_{II} = \gamma_o\left[1 - (\sigma/2)\sin^2(2\beta)\right]$

and

$\gamma_{\perp} = -\gamma_o(\sigma/4)\sin(4\beta)$

with

$\gamma_o = (6\pi/\lambda n)\chi^{(3)}_{xxxx}$, β is the angle of the input polarization plane with respect to the [100] axis of the crystal

and

$$\sigma = \frac{\chi^{(3)}_{xxxx} - \left(2\chi^{(3)}_{xyxy} + \chi^{(3)}_{xxyy}\right)}{\chi^{(3)}_{xxxx}}$$

which is the anisotropy of the $\chi^{(3)}$ tensor. In general case γ_{II} and $\gamma_{\perp}$ are complex: $\gamma_{II} = \gamma'_{II} + i\gamma''_{II}$ and $\gamma_{\perp} = \gamma'_{\perp} + i\gamma''_{\perp}$. The solution to system (1) with as initial conditions $B(0) = 0$ and $A(0) = A_o$ (A_o real) is:

$$A = A_o \exp\left(i\gamma_{II} A_o^2 L\right), \tag{29a}$$

$$B = A_o\left(\gamma_{\perp}/\gamma_{II}\right)[\exp(i\gamma_{II} A_o^2 L) - 1]. \tag{29b}$$

The complex amplitudes B and A define the ratio $R = B/A$. The angle position δ_o of the main ellipse axis and the ellipticity angle ε can be found using the relations $\tan(2\delta_o) = 2\,\mathrm{Re}(R)\Big/\left(1-|R|^2\right)$ and $\sin(2\varepsilon) = 2\,\mathrm{Im}(R)\Big/\left(1+|R|^2\right)$, already presented in (15a) and (15b). Assuming δ_o, $\gamma'_{II} A_o^2 L, \gamma''_{II} A_o^2 L$, $\gamma'_{\perp} A_o^2 L$ and $\gamma''_{\perp} A_o^2 L$ small we get:

$$\delta_o \approx \frac{1}{2}\gamma'_{\perp}\gamma'_{II}\, A_o^4 L^2\,, \tag{30}$$

$$\varepsilon^2 \approx \left(\gamma'_{\perp} A_o^2 L\right)^2. \tag{31}$$

The expression (31) indicates that the magnitude of XPW efficiency is proportional to the square of the input intensity in accordance with previous understanding of this effect. However the relation (30) predicts that polarization rotation depends on the square of the input intensity and on the real part of the cubic nonlinearity. This is in contrast to the previously reported theoretical and experimental investigations obtained at relatively lower intensity and bigger contribution of the two photon absorption effects.

The experiment (Minkovski *et al.* 2002) with the cubic crystal yttrium vanadate (YVO_4) crystal confirmed the theoretical prediction in (30). For studying the output polarization state extinction curve measurements were performed by introducing small angular deviations δ from the exact crossed position between the two polarizers. For analyzing sign of ellipticity, a $\lambda/4$ quartz plate with axis c parallel to input polarization is introduced in front of analyzer. The maximum XPW efficiency achieved is 1.2% .

Typical results of extinction curve measurements are shown on Fig 10 for two input intensities. The solid lines are parabolic fits of the type $Y = C_1(\delta - \delta_o)^2 + C_2$. The parameter δ_o is a measure of the induced polarization

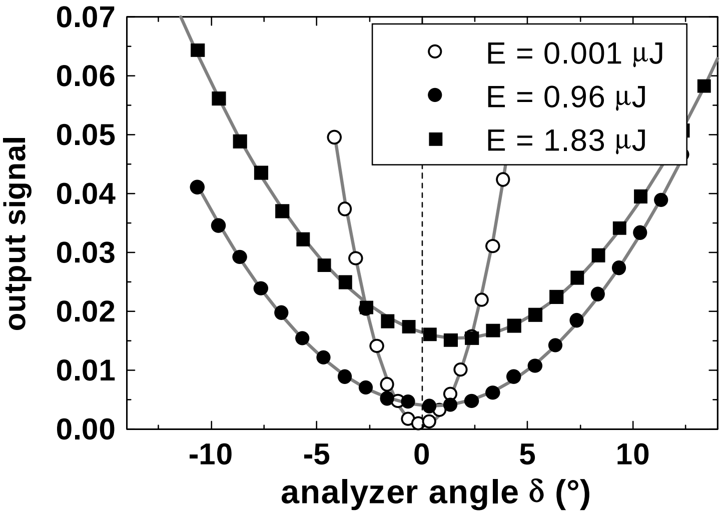

Figure 10 . *Output signal (normalized to input intensity) with respect to angular detuning of the analyzer for three different input energies. Zero position corresponds to exactly crossed polarizer and analyzer. Solid lines are parabolic fit.*

rotation angle and $|\varepsilon| = \sqrt{C_2/C_1}$, a measure of the modulus of input ellipsity. For the curve recorded at input energy 0.96 μJ the fitting gives $\delta_o = +8.7$ mrad and $|\varepsilon| = 64$ mrad, and for the curve recorded at input energy 1.83 μJ the fitting gives $\delta_o = +30$ mrad and $|\varepsilon| = 122.5$ mrad. Additional test with $\lambda/4$ plate proved that $\varepsilon < 0$. As seen from figure 10 the dependence of δ_o on intensity measured for three values of input intensity is quadratic in good accordance with the theory described above. We measured the amount of anisotropy of two-photon absorption and estimated that induced rotation due to two-photon absorption can not explain the recorded polarization rotation angles.

From Equation (30) we see the role of the cascade processes. The expression for induced polarization rotation angle δ_o is proportional to $\chi^{(3)}:\chi^{(3)}$ term. The reason for this "cascaded rotation" lies in the self-phase modulation effect experienced by the fundamental wave A. At high input intensities the phase shift between B and A will be different from $\pi/2$ and intensity dependent: $\arg(R) = (\pi - \gamma_{II} A_o^2 L)/2$. This leads to complex value of Γ and consequently to induced polarization rotation. It is the «cascading» of self-phase modulation of the fundamental wave and generation of cross-polarized wave by four-wave mixing that causes the induced polarization rotation. The described effect can be observed along crystallographic axes of cubic crystals or along Z axis of tetragonal crystals. The condition for observing the effect is $\gamma''_\perp$ to be very small in combination with relatively high intensity, so that $\gamma''_\perp / \left(\gamma'_\perp \gamma'_{II} A_o^2 L\right) << 1$. This condition is easy to satisfy when fundamental photon energy is below half of the band gap. In our experiment the fundamental photon energy is near the edge of the half of the band gap, but the above inequality is well fulfilled due to high intensity used in the described experiment.

The effect of $\chi^{(3)}:\chi^{(3)}$ cascaded induced rotation of the polarization is unique. This is not just a simple mimic of $\chi^{(5)}$ contribution - the effect of "cascaded" nonlinear polarization rotation reported here has no $\chi^{(5)}$ analogue.

The knowledge about this effect is important for avoiding unwanted depolarization effects. For example, YVO_4 is known as an active element for solid state lasers. Also this effect can be used for measuring the magnitude and sign of the components of $\chi^{(3)}$ tensor and therefore the anisotropy of the $\chi^{(3)}$ tensor. The attractive aspect of such a method lies in the single beam technique with pumping and recording at one and the same wavelength and that the measurement is based on recording angle rotation values. On the other hand, if the cubic nonlinearity is known, then measurement of the induced polarization rotation angle can be used for calculating the intensity of the beam. Finally polarization-switching devices, can also be designed.

The common characteristic of considered in part III interactions is that they consist several steps, each of them with own phase-matching condition. For best efficiency simultaneous phase matching of all steps is required. Some of the interactions (e. g. cascaded frequency conversion and the effect of cascaded polarization rotation in centosymmetric media) does not require special effort for fulfilling double phase-matching. Double phase matching is achieved automatically. For realization of the other interactions like, third harmonic, fourth harmonic and XPW generation in single quadratic crystal require the nonlinear media to be prepared in a special way for supporting double or multiple phase matching conditions. The methods for achievement of double phase-matching is considered in the next part.

IV. Methods for achievement of simultaneous phase-matching for several processes

In this part we present a brief overview of different methods for simultaneous phase-matching of several parametric optical processes (the so-called *multistep cascading*) in engineered structures with the modulated second-order nonlinear susceptibility. In particular, we discuss the possibility of double phase-matching (DPM) in both uniform and non-uniform quasi-phase-matched (QPM) periodic optical superlattices and also in the recently fabricated two-dimensional nonlinear photonic crystals.

IV.1. DPM with uniform QPM structures –method of two commensurable periods.

In a bulk homogeneous nonlinear crystal the quadratic nonlinearity is constant everywhere. Several methods (Fejer *et al.* 1992) have been suggested and employed in order to create a periodic change of the sign of the second-order nonlinear susceptibility $d^{(2)}$ in QPM structures as shown in Figure 11a. From the mathematical point of view, such a periodic sequence of two domains can be described by the periodic function

$$d(z) = d_o \sum_{m \neq 0} g_m \exp(iG_m z), \quad (32)$$

$$g_m = (2/m\pi)\sin(\pi m D), \quad (33)$$

where $G_m = 2\pi m/\Lambda$ is the m-th reciprocal vector of the QPM structure. The uniform QPM structure is characterized by a set of the reciprocal vectors: $\pm 2\pi/\Lambda$, $\pm 4\pi/\Lambda$, $\pm 6\pi/\Lambda$, $\pm 8\pi/\Lambda$, ..., which can be used to achieve the phase-matching conditions provided. The princip how the reciprocal vector compencate the mismatch of a given nonlinear process is shown on figure 11b, where the process of sum-frequency mixing is taken as example. The integer number *m* (that can be both *positive* or *negative*) is called *the order of the QPM phase-matching*. According to Eq. (33), the smaller is the order of the QPM

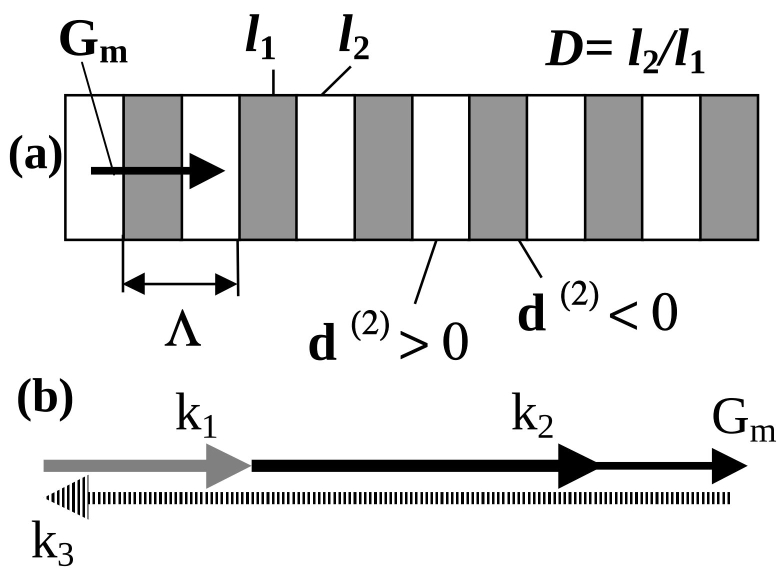

Figure 11. *QPM structure (a) and the princip (b) of phase-matching compensation with the help of the grating reversal vectors*

reciprocal wave vector, the larger is the effective nonlinearity. If the filling factor D = 0.5, the effective quadratic nonlinearities (proportional to the parameter $d_o g_m$) that correspond to the even orders QPM vectors vanish. Importantly, such uniform QPM structures can be used for simultaneous phase-matching of two parametric processes (Pfister *et al* 1997).

As an example, we consider the cascaded third harmonic in single crystal under the condition that all interacting waves are *collinear* to the reciprocal wave vectors of the QPM structure. We denote the mismatches of the nonlinear material without modulation of the quadratic nonlinearity ("bulk mismatches") as Δb_1 and Δb_2 (i.e., $\Delta b_1 = k_2 - 2k_1$ and $\Delta b_2 = k_3 - k_2 - k_1$) and choose the period Λ of the QPM structure in order to satisfy the phase-matching conditions $G_{m_1} = -\Delta b_1$ and $G_{m_2} = -\Delta b_2$. Two processes, SHG and SFM, are characterized by the wave-vector mismatches $\Delta k_1 = k_2 - 2k_1 + G_{m_1} = 0$ and $\Delta k_2 = k_3 - k_2 - k_1 + G_{m_2} = 0$, respectively, and they become simultaneously phase-matched for this particular choice of the QPM period. A drawback of this method is that it can satisfy simultaneously two phase-matching conditions for *discrete values* of the optical wavelength only. The values of the fundamental wavelength λ for the double phase-matching condition can be found from the relation

$$\Delta b_2/m_2 - \Delta b_1/m_1 = 0, \qquad (34)$$

since both Δb_1 and Δb_2 are functions of the wavelength. For a chosen pair of integer numbers (m_1, m_2) the QPM period Λ is found from the relation $\Lambda = 2\pi|m_1/\Delta b_1|$ or $\Lambda = 2\pi|m_2/\Delta b_2|$. Some tunability is possible due to the dependence of Δb_1 and Δb_2 on temperature.

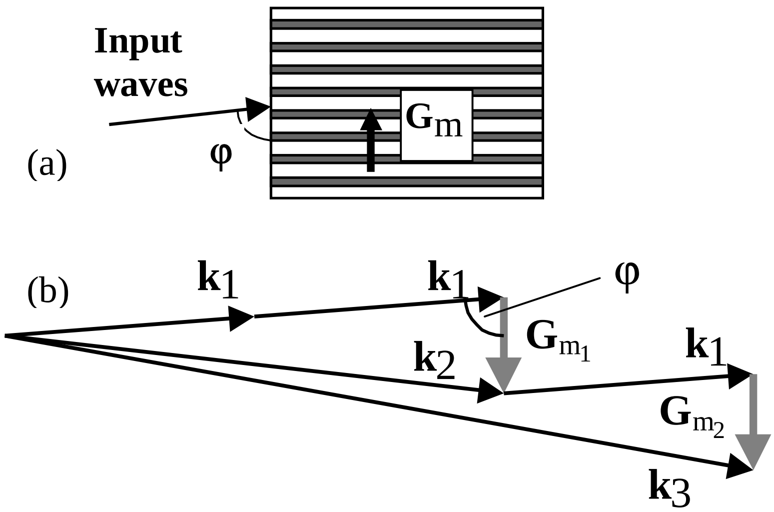

Figure 12. *Noncollinear interaction with QPM structure (a) and the princip (b) of double-phase-matching for the third harmonic generation via sum frequency mixing of fundamental and second harmonic wave*

IV.2. DPM with uniform QPM structures –noncollinear case.

Phase-matching is possible even in the case when some of the waves propagate under a certain angle to the direction of the reciprocal vectors of the QPM structure, i. e. for the non-collinear case. The advantage of this method is that double phase-matching can be realized in a broad spectral range. Such a type of noncollinear interaction will be efficient for the distances corresponding to the overlap of the interacting beams.

Let us consider the cascaded single crystal third harmonic generation process in this type of geometry (see Figure 2). As has been discussed above, the simultaneous phase-matching of two parametric processes $\omega + \omega = 2\omega$ and $\omega + 2\omega = 3\omega$ is required in this case. We assume the fundamental wave at the input. As shown in Figure 12b, for the first process, SHG, the phase-matching is achieved by the reciprocal vector $\mathbf{G}_{m_1}$ and the generated SH wave is with wave-vector $\mathbf{k}_2$

not collinear to the fundamental wave: $2\mathbf{k}_1 + \mathbf{G}_{m_2} = \mathbf{k}_2$. The second process, sum frequency mixing, is phase-matched by the vector $\mathbf{G}_{m_2}$: $\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{G}_{m_2} = \mathbf{k}_3$. From Figure 12 we can calculate the period of the QPM structure that allows achieving double phase-matching of two processes - SHG and sum frequency mixing (Saltiel and Kivshar 2000a):

$$\Lambda = 2\pi \sqrt{\frac{2(k_3^2 - 9k_1^2)m_1 - 3(k_2^2 - 4k_1^2)(m_1 + m_2)}{(m_1 + m_2)(2m_2 - m_1)m_1}} \tag{35}$$

Because of the two degree of freedom: the angle of noncollinearity φ and the QPM period Λ, the double-phase matching can be realized in wide spectral range.

IV.3. DPM with non-uniform QPM structures

Several nonuniform QPM structures have been proposed and experimentally tested. One type is so called phase-reversed QPM structure. The idea of the phase-reversed QPM structures (Chou *et al* 1999) is illustrated in Fig. 13a. Such a structure can be described as a sequence of many equivalent uniform short QPM sub-structures with the length $\Lambda_p/2$ connected in such a way that at the place of the joint two end layers has the same sign of the quadratic nonlinearity. Any two neighboring junctions have opposite sign of the $\chi^{(2)}$ nonlinearity. By other words, the phase-reversed QPM structure is a kind of an uniform QPM structure with a change of the domain phase by π characterized by the other (larger) period Λ_p.

The modulation of the quadratic nonlinearity $d(z)$ in the phase-reversed QPM structure with the filling factor $D = 0.5$ can be described by the response function

$$d(z) = d_o(-1)^{\mathrm{int}(2z/\Lambda_Q)}(-1)^{\mathrm{int}(2z/\Lambda_p)} \tag{36}$$

and can be presented in the form of the Fourier series,

$$d(z) = d_o\left[\sum_{-\infty}^{+\infty} g_l e^{(iG_l z)}\right]\left[\sum_{-\infty}^{+\infty} g_m e^{(iF_m z)}\right] = d_o\left[\sum g_{lm} e^{(iG_{lm} z)}\right], \quad l, m \neq 0, \tag{37}$$

where $g_m = \dfrac{2}{m\pi}$, $g_{lm} = g_l g_m$, $G_l = \dfrac{2\pi l}{\Lambda_Q}$, $F_m = \dfrac{2\pi m}{\Lambda_p}$, $G_{lm} = \dfrac{2\pi}{\Lambda_Q} l + \dfrac{2\pi}{\Lambda_p} m$.

Another type non-uniform QPM structure is proposed by Bang *et al.* (1999). This structure is illustrated in fig. 13b and is characterized by the QPM period Λ_Q that is a periodic function of z:

$$\Lambda = \Lambda_Q + \varepsilon_o \cos(2\pi z/\Lambda_p). \tag{38}$$

The formulas for double phase-matching are the same for the both phase reversed and periodically chirped QPM structures shown on figure 13.

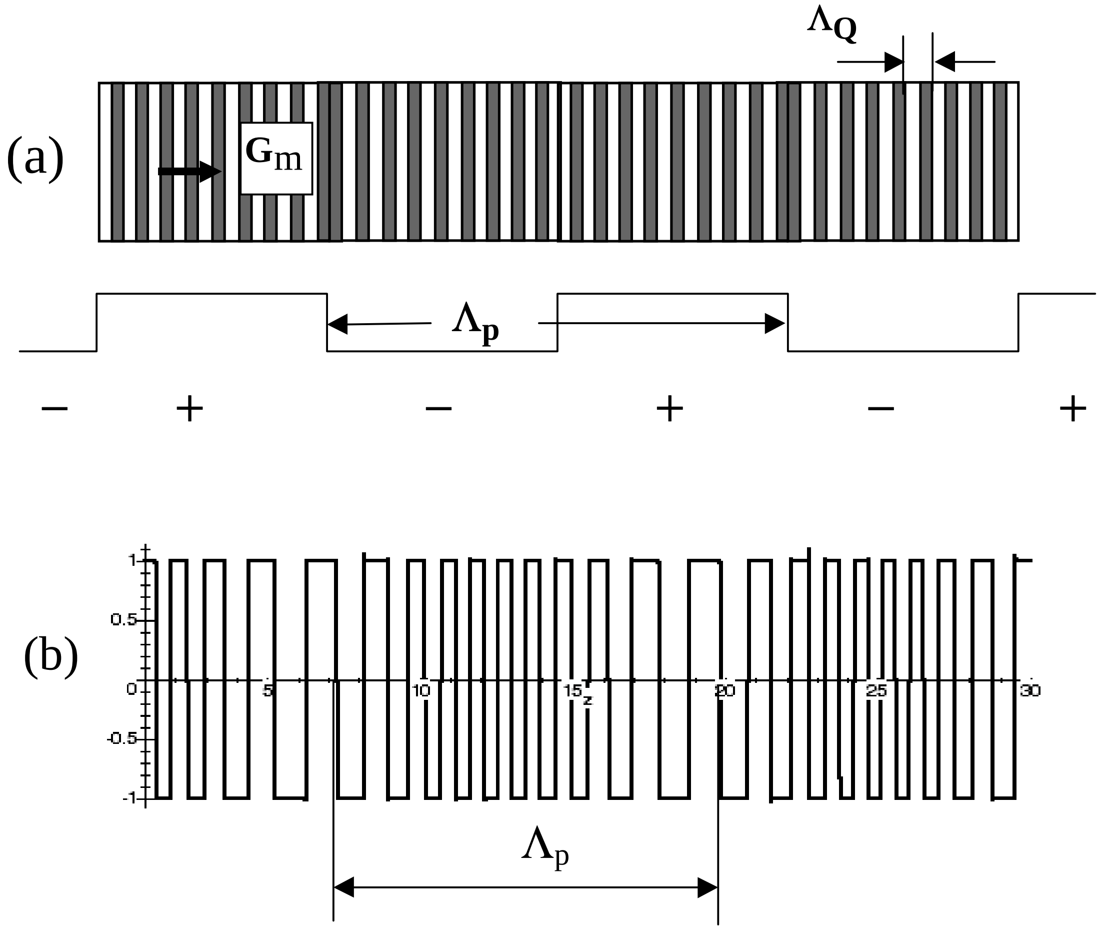

Figure 13. *Non-uniform QPM structures: (a) phase reversed QPM structure (*Chou *et al* 1999*) and (b) periodically chirped QPM structure (*Bang *et al.* 1999*).*

$$\Lambda_Q = \left| \frac{2\pi (l_2 m_1 - l_1 m_2)}{m_2 \Delta b_1 - m_1 \Delta b_2} \right| \quad , \tag{39}$$

$$\Lambda_p = \left| \frac{2\pi (l_2 m_1 - l_1 m_2)}{l_2 \Delta b_1 - l_1 \Delta b_2} \right| \quad . \tag{40}$$

Equations (39,40) allow the two periods of these non-uniform QPM structures to be found. The designed this way structures will support double phase matching of any two nonlinear optical processes with bulk phase mismatches Δb_1 and Δb_2. In very recent works (Liu *et al.* 2001,2002) demonstrated that phase-reversed structures are really suitable for single crystal THG.

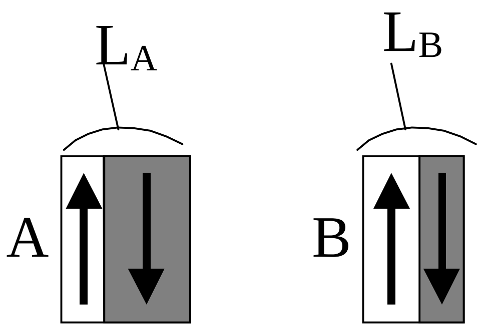

Figure 14. *The two basic blocks for construction quasi-periodic and aperiodic optical superlattices*

IV.4. DPM with quasi-periodic and aperiodic optical superlattices

Another method of double-phase-matching, investigated both theoretically and experimentally is based on the use of the quasi-periodic optical superlattices (QPOS) (Zhu and Ming 1999) and aperiodic optical superlattices (Zhang *et al* 2000). In the most of the cases QOPSs are built with two-component blocks (A and B) aligned in a Fibonacci (or more general quasi-periodic) sequence (see Fig. 14). Each of the blocks consists of two layers with the opposite sign of the quadratic nonlinearity. Then the two blocks are aligned in Fibonacci (ABAABABA….) or different sequence. We illustrate the possibility of double-phase-matching in such a kind of structures taking as an example, the structure that consists two blocks, aligned in a generalized sequence (Fradkin-Kashi *et al.* 1999, 2002). The modulation of the quadratic nonlinearity $d(z)$ can be described by the following Fourier expansion:

$$d(z) = d_o \sum_{m,n} f_{m,n} \exp\left(iG_{m,n}z\right) , \qquad (41)$$

where the reciprocal vectors are defined as $G_{m,n} = 2\pi\left(m + n\tau\right)/S$ and $S = \tau L_A + L_B$.

For phase-matching of two processes with bulk mismatches Δb_1 and Δb_2, we solve the system of equations

$$G_{m_1,n_1} = 2\pi\left(m_1 + n_1\tau\right)/S = \Delta b_1 , \qquad (42a)$$

$$G_{m_2,n_2} = 2\pi\left(m_2 + n_2\tau\right)/S = \Delta b_2 \qquad (42b)$$

and find unknown *S* and τ. Exact values for L_A, L_B and the lengths of the layers have to be found by maximizing f_{m_1,n_1} and f_{m_2,n_2}. This structure allows simultaneous phase-matching in a broad spectral range without constrains on the ratio $\Delta b_2 / \Delta b_1$. Equations (42) are also valid for Fibonacci type QPOS but, since τ is fixed, double phase-matching can be satisfied for a limited number of the wavelengths.

In several recent papers aperiodic optical superlattices were used for simultaneous phase- matching of several processes. Presently the record efficiency in single crystal THG with the use of generalized two-component QPOS is 27% (Zhang *et al* 2001).

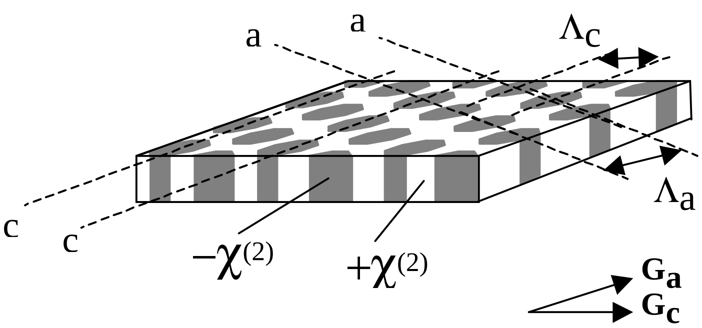

Figure 15. *2D Nonlinear photonic crystals is a periodic lattice of volumes (marked in gray) with reversed sign of the* $\chi^{(2)}$. $\mathbf{G_a}$ *and* $\mathbf{G_c}$ *are the reciprocal vectors of the 2D structure.*

IV.4. Two dimensional $\chi^{(2)}$ nonlinear photonic crystals as environment for DPM

Nonlinear photonic crystals (NPC) with homogeneous (linear) refraction index and two dimensional (2D) periodic lattice of volumes with reversed sign of the quadratic nonlinear susceptibility can be effectively employed as a host media for many different types of CNOP (Berger 1998). A schematic diagram of 2D NPC is shown in Fig. 15. The simple way to obtain the phase-matching conditions for 2D NPC is to use a reciprocal lattice formed by vectors $\mathbf{G_a}$ and $\mathbf{G_c}$, defined as $|G_a| = 2\pi/\Lambda_a$ and $|G_c| = 2\pi/\Lambda_c$ and directions - the normals to the lines **a** and **c**, respectively . Vectors $\mathbf{G_a}$ and $\mathbf{G_c}$ are used to construct so call reciprocal lattice, that is very useful for analyzing phase matching conditions in these type photonic structures.

For a hexagon lattice, we find $\Lambda_a = \Lambda_c = a\sqrt{3}/2$, where a is the distance between the centers of two neighboring volumes with inverted sign of $\chi^{(2)}$ and . For asymmetric unit cell $\Lambda_a \neq \Lambda_c$ and the two basic reciprocal vectors $\mathbf{G_a}$ and $\mathbf{G_c}$ has different modulus that depends on the lattice spacing a, asymmetry ε and the angle δ of the unit cell (Saltiel and Kivshar 2000b). All reciprocal vectors of the 2D NPC crystal are formed by a simple rule, $\mathbf{G}_{m,n} = m\mathbf{G}_c + n\mathbf{G}_a$. Any two vectors of this set $\{\mathbf{G}_{m,n}\}$ can be used to compensate for the bulk mismatches Δb_1 and Δb_2; however in most of the cases such phase-matching conditions assume noncollinear interactions. Phase-matching conditions for single crystal cascade third harmonic generation process are shown in Fig. 16: (a) the first step $\omega + \omega = 2\omega$ is phase-matched by the reciprocal vector $\mathbf{G}_{m_1,n_1}$ and (b) the second step $\omega + 2\omega = 3\omega$ is phase-matched by the reciprocal vector $\mathbf{G}_{m_2,n_2}$. Phase-matching is achieved by choosing the lattice spacing a and the angle of incidence β. 2D NPC can be also used for simultaneously phase-matching *of three nonlinear processes,* e.g. second, third, and fourth harmonic generation (Saltiel and Kivshar 2000b) or generation of pair SH waves and fourth harmonic generation (de Sterke *et al* 2001). Experimentally, the simultaneous second-, third-, and fourth-harmonic generation was recently observed in poled $LiNbO_3$ 2D NPC (Broderick *et al* 2000).

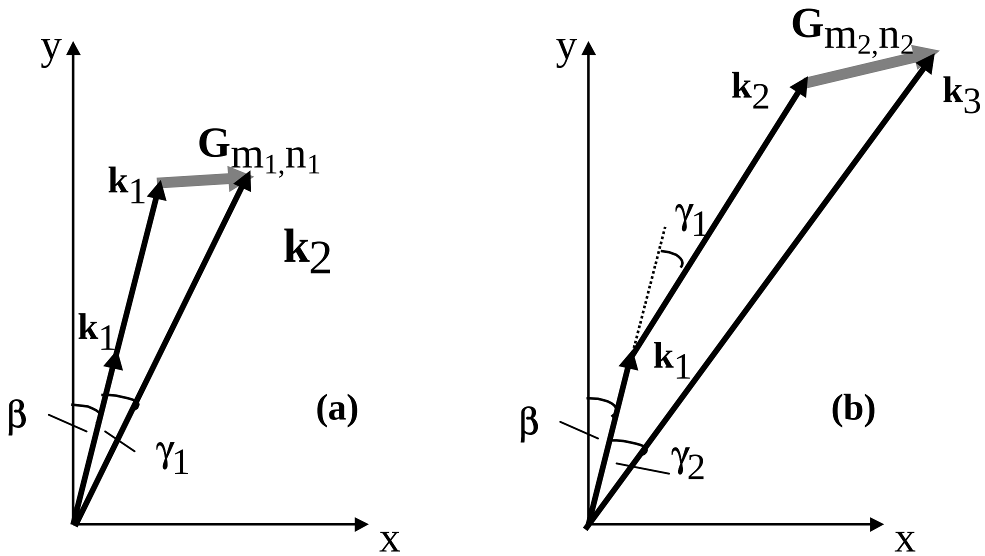

Figure 16. *Double phase-matching for single crystal third harmonic in 2D NPC: (a) the process $\omega + \omega = 2\omega$ is phase-matched by the reciprocal vector $\mathbf{G}_{m_1,n_1}$ and (b) the process $\omega + 2\omega = 3\omega$ phase-matched by the reciprocal vector $\mathbf{G}_{m_2,n_2}$.*

V. Examples of cascading of second order processes in 2D NPC

V.1. Collinear third and four harmonic generation in 2D NPC

In general case when nonlinear process is realized in 2D NPC, as shown above, all interaction waves are non-collinear. When the input beam(s) are with finite width the noncollinearity of the interaction leads to reduced efficiency. However with special design of the unit cell it is possible to construct of cascaded interactions in which the final generated wave is collinear to the input fundamental wave. Such interactions for four harmonic generation (de Sterke *et al.* 2001) and third harmonic generation (Karaulanov and Saltiel 2002) have been proposed. Both proposed interactions starts with two channels SHG that propagate at small angles at both sides of the direction of the fundamental wave. The possibility for two channel harmonics generation is characteristic only for 2D NPC, when the unit cell is symmetrical (i.e. asymmetry $\varepsilon = 1$ **).** For this kind of 2D NPC at certain directions $\left|\mathbf{G}_{m,\,n}\right| = \left|\mathbf{G}_{m,-n}\right|$ and when these directions are used for the propagation of the fundamental beam two channel SHG can be generated. The idea for collinear third and fourth harmonic generation in 2D NPC is clarified in Figure 17. The two channel SHG results in for times higher efficiency for the process of SHG (in comparison with single channel and suggesting the same nonlinearity). The higher efficiency for the SH process results in higher efficiency for total process of four harmonic generation. As seen from Figure 17 the second step of the suggested scheme for third harmonic generation is also “two channeled”, that should lead to

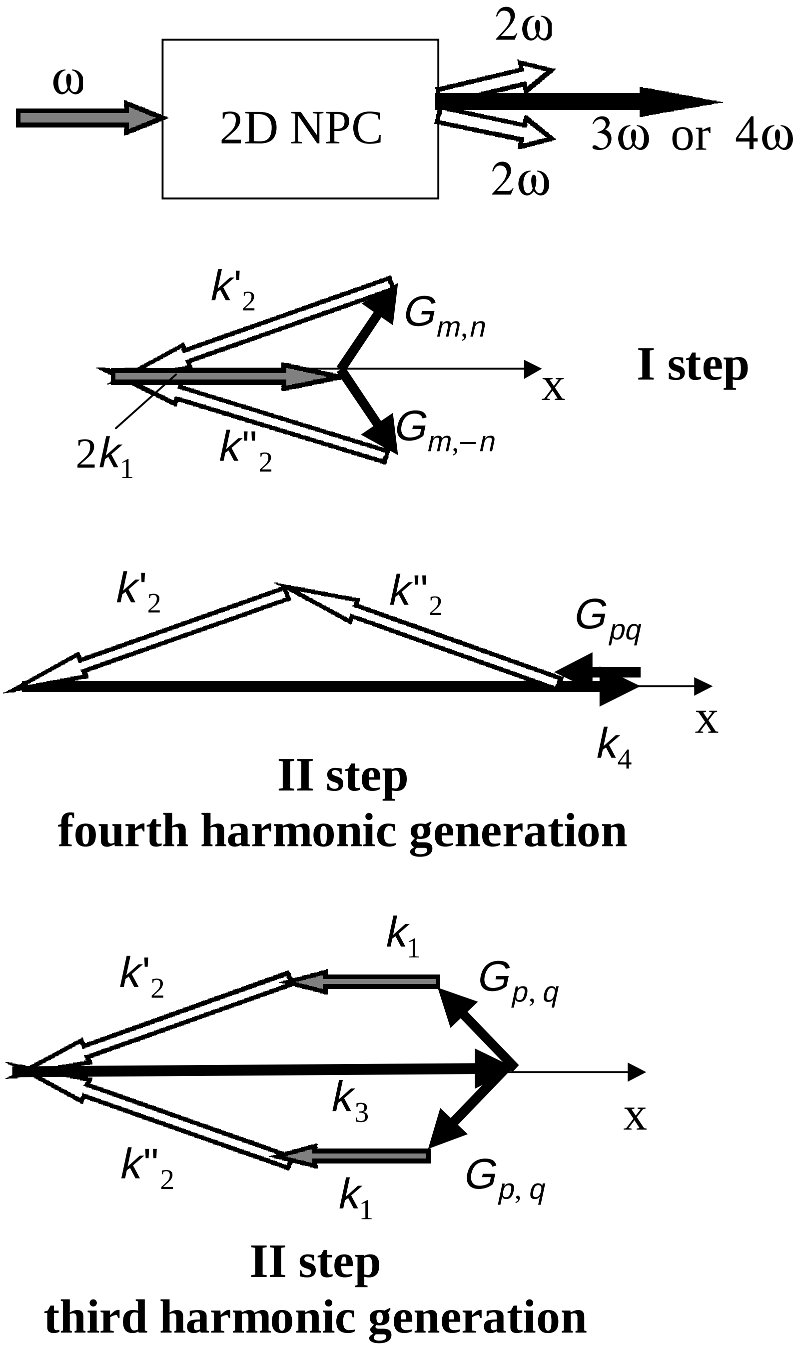

Figure 17. *Double phase-matching schemes for generation of collinear third and four harmonic in single 2D NPC*

increased efficiency. Indeed, for the process of four harmonic generation in assumption of nondepletion of the SH wave due to generated fourth harmonic wave one can obtain for the overall efficiency (de Sterke *et al*. 2001)

$$\eta_{4\omega} = \sigma_2^2 \left[A_0 L - \frac{1}{\sqrt{2}\sigma_1} \tanh\left(\sqrt{2}\sigma_1 A_0 L\right) \right]^2 , \tag{43}$$

where σ_1 and σ_2 are the two coupling coefficient that control the first and the second step. For comparison, the result for the single-channel case can be obtained from Eq. (43) by replacing $\sqrt{2}\sigma_1$ with σ_1.

V.2. Wavelength routing with interchange of the information.

The noncollinearity as a characteristic for the interactions in 2DNPC can be useful for interactions where splitting of several information channels is required. Such an example is the reported experimental investigation of wavelength routing with interchange of the information (Chowdhury *et al* 2000, 2001).

In the proposed scheme two information signals one at wavelength ω_a (modulated with information $\mathbf{I}_1$) and second - at ω_b (modulated with information $\mathbf{I}_2$) and pump beam enter 2D NPC. As a result of two simultaneously phase matched second order frequency mixing processes at the output of the crystal two deflected signals will be generated at the same frequencies, but with exchanged information: output deflected signal at wavelength ω_a is modulated with information $\mathbf{I}_2$ and output deflected signal at wavelength ω_b is with information $\mathbf{I}_1$. The balance of the wave-vectors and the frequencies is given by :

$$\begin{aligned} \mathbf{k}_p - \mathbf{k}_a &= \mathbf{k}'_b + G_{ij} \\ \mathbf{k}_p - \mathbf{k}_b &= \mathbf{k}'_a + G_{mn} \end{aligned} ; \tag{44}$$

$$\begin{aligned} \omega_p - \omega_a &= \omega_b \\ \omega_p - \omega_b &= \omega_a \end{aligned} , \tag{45}$$

where "prime" indicates the wave-vectors of the output signal waves and and G_{ij} and G_{mn} are the two reciprocal vectors that phase match the two interactions.

The equations that describe these double phase-matched interaction are:

$$\begin{aligned} \frac{dA_1}{dz} &= -i\sigma P B^* \qquad & \frac{dA}{dz} &= -i\sigma P B_1^* \\ \frac{dB_1}{dz} &= -i\sigma P A^* \qquad & \frac{dB}{dz} &= -i\sigma P A_1^* \end{aligned} \tag{46}$$

The solution of this system gives not only exchange of the information, but also amplification of the two output deflected signals (A_1, B_1) and depletion (T) of the output nondeflected signals (A , B):

$$\eta_{A1} = (A_1/B_o)^2 = \eta_{B1} = (B_1/A_o)^2 = \sinh^2(\sigma P x) \tag{47}$$

$$T_A = (A/A_o)^2 = T_B = (B/B_o)^2 = \cosh^2(\sigma P x) \tag{48}$$

V.3. All-optical deflection by second-order cascading in 2D NPC

In our last example we will consider the possibility of all optical pump induced deflection of signals in 2D NPC (Saltiel and Kivshar 2002). The idea is shown in Figure 18a. Input pump and probe are at the same wavelength and are polarized in perpendicular planes. The result of cascaded interaction is that the signal is deflected only when the strong pump is present. In this sense this device can be called nonlinear optical polarizer that is controlled by pump. The cascading interaction includes two steps: in the first one the pump is frequency doubled with a process that is phase matched with a reciprocal vector collinear to the pump wave-vector. Generated SH wave collinear with the pump mixes with the input signal (also collinear with the pump) generating a deflected signal at the same frequency and polarized the same way as the input one. The reason for the deflection is that phase matching with achieved with arbitrary oriented reciprocal vector. The balance of the wave-vectors and the frequencies for the two discussed steps is given by:

$$\begin{aligned} &\omega_p + \omega_p = 2\omega_p = \omega_2 \\ &\omega_2 - \omega_s = \omega_s' \end{aligned} \tag{49}$$

$$\begin{aligned} &\mathbf{k}_p + \mathbf{k}_p = \mathbf{k}_2 + \mathbf{K}_{ij} \\ &\mathbf{k}_2 = \mathbf{k}_s + \mathbf{k}_s' + \mathbf{K}_{mn} \end{aligned} \tag{50}$$

The amplitude of the deflected and transmitted signal is given by

$$\eta_s(\eta_{s'}) = \frac{1}{4}\left\{ \left[\operatorname{sech}(\sigma_1 A_p L)\right]^{\left|\frac{\sigma_2}{\sigma_1}\right|} \pm \left[\cosh(\sigma_1 A_p L)\right]^{\left|\frac{\sigma_2}{\sigma_1}\right|} \right\}^2 \tag{51}$$

where $\eta_s = [A_s(L)/A_s(0)]^2$, A_o is the input pump's amplitude, $A_s(0)$ is the input signal's amplitude, L is the crystal length; σ_1 and σ_2 are the two coupling coefficient that control the first and the second step ; signs "+" and "-" correspond to transmission efficiency of the nondeflected and deflected output signals, respectively. The obtained with expressions (51) dependenceis are shown on Fig. 18b. It is seen that both deflected and nondeflected signals at certain levels of the pump and length of the media will be amplified.

VI. Conclusion

In conclusion, in this chapter a systemization and examples of cascaded nonlinear optical processes are presented. One part of cascaded interactions, that are controlled by single phase mismatch parameter, are responsible for self interaction effects like self modulation, induced polarization rotation, soliton propagation, compression and etc. Respectively, such kind of interactions can be used for construction of devices for routing, all-optical switching, mode locking and others.

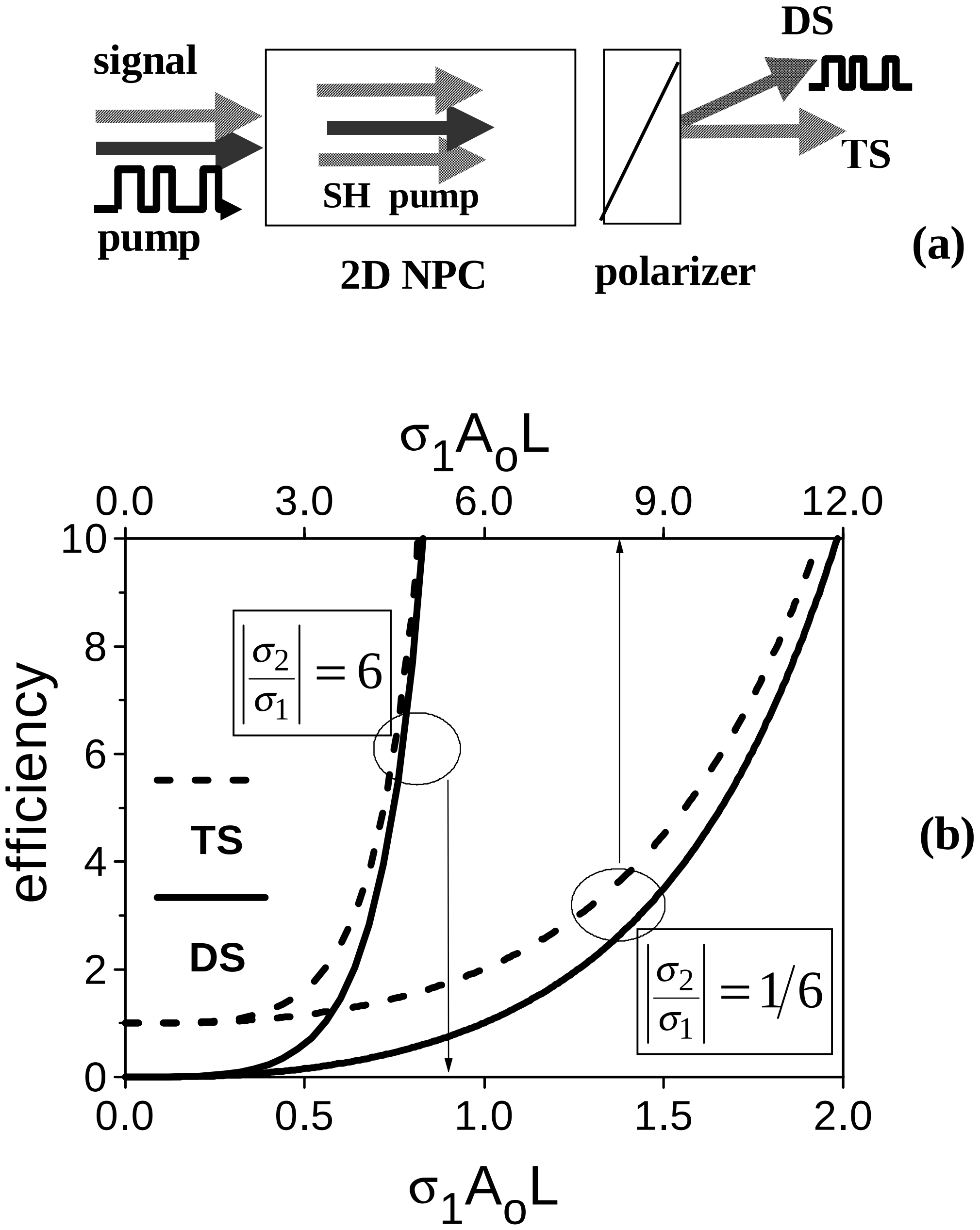

Figure 18. *(a) Schematic of the suggested cascading interaction for deflection in 2D NPC. Signal and pump are cross polarized. TS, transmitted signal; DS, deflected signal.*
(b) Efficiency of the deflected (DS) and non-deflected (TS) signals relative to normalized input pump amplitude the deflection scheme and for two values of the ratio σ_2/σ_1 .

The second group CNOP are those that are controlled by two phase-matched parameters. These types of interactions are mainly responsible for generation of new waves in single nonlinear media. For example, recently the interest toward third harmonic generation in single quadratic crystal is extremely strong due to the tendency of constriction of compact diode-pumped femtosecond systems with integrated single crystal frequency convertors in UV range. Other applications of the double-phase matched interactions are fourth harmonic generation and frequency conversion for optical communication, cross-polarized wave generation. For best efficiency both phase matched conditions should be satisfied. The review of the methods for achievement of simultaneous phase-matching is also presented in this chapter. In several experimental works it is already proved that phase reversed QPM grating, quisi-periodical optical superlattices and two-dimensional nonlinear photonic crystals are suitable structures that can be predesigned for achievement of double phase matching. Several promising cascaded interaction that can exist only in two-dimensional nonlinear photonic crystals have been discussed.

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