Influence of the Spectral Phase on the Efficiency and the Spectral Output Parameters in the Processes of Second Harmonic and Cross-Polarized Wave Generation∗

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Abstract. We derive the main equations that describe the evolution of the spectral properties for two different nonlinear optical processes: second harmonic (SH) generation as representative of the quadratic processes and cross polarized wave (XPW) generation as representative of third order nonlinear optical processes. The role of both second order and third order spectral phases on the output parameters of the SH wave and the XPW are studied. The approach can be applied to other nonlinear optical processes as well.

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1 Introduction

For description of the processes in nonlinear optics we can use time domain and spectral domain approaches. Most frequently is used time domain approach but spectral domain approach very often offers other attractive opportunities like easier analytical representation of the processes when we use ultrashort light pulses, simple description of frequency shift spectral shapes and the other parameters of the converted spectrum. We will note that spectrum of ultrashort femtosecond pulse is measured much easier than time shape of the same pulse. For this reason we have chosen the spectral approach to describe the output parameters for two popular nonlinear processes: second harmonic generation and cross polarized wave generation.

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†The paper was received a couple of weeks before Prof. Solomon Mois Saltiel, an elected member of the Bulgarian Academy of Sciences and member of the editorial board of the Bulgarian Journal of Physics, suddenly passed away.
Second harmonic generation (SHG) is the most popular second order nonlinear process that finds application in science and technology. With the progress in the field of generation shorter and shorter pulses in the femtosecond range became actual to obtain more information about how the input spectral phases influence the efficiency of the SHG process and whether output spectral phases are enhanced or reduced during the SHG process. Some preliminary results concerning the second order spectral phase are considered in [1]. In this paper we derive the equations describing the transformation of the spectral properties of the fundamental waves and for the first time we present an analysis of the role of third order spectral phase on the efficiency of the SHG process, on the SH spectral width and shift.

The Cross-Polarized Wave (XPW) generation is a third order process in which a new wave polarized in direction perpendicular to the input one is efficiently generated in oriented cubic crystal [2]. In this process input and output waves are at the same frequency, but the output wave has different orientation of the polarization plane. This process was known a long time ago [3], but only recently it has been realized that the efficiencies up to 30% can be achieved (see e.g. [4]) and also that XPW generation process can have very important applications as for example contrast enhancement of femtosecond pulses (see [5]) and the references there. As XPW generation is a third order nonlinear optical process, it also modifies the femtosecond pulse itself. The effect of a pure second order phase on the XPW spectrum has been discussed in [6]. It is shown there that in the case of zero second order phase the XPW spectrum is broader by a factor $\sqrt{3}$ compared to the initial one. The role of third and higher order phase terms has been studied in [7]. Here we extend this study by giving detailed derivation of the equations describing the modification of the spectral properties of the input pulse during this cubic process. We present for the first time the output spectral shapes as a function of the input spectral phases, demonstrating that both blue and red shift of the SH spectrum can be expected depending on the exact values of the input spectral phases.

2 Spectral Approach for SHG and XPW Generation

In the equations we are going to derive we consider the approximation of non-depleted fundamental wave, which means that we can consider the fundamental wave as propagating in linear medium. For this reason we have to remind the fundamental for the spectral shape of a wave propagating in linear medium.

2.1 Fundamental Wave Equation

Let us start considering propagation of the fundamental (input) laser wave $A(z, t)$ in a medium without nonlinearity

$$\frac{\partial A(z, t)}{\partial z} + \frac{1}{u} \frac{\partial A(z, t)}{\partial t} - \frac{k_2}{2} \frac{\partial^2 A(z, t)}{\partial t^2} = 0,$$

(1)
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where \( u \) is the group velocity of the wave and \( k_2 \) is the second order dispersion coefficient. It is convenient to rewrite Eq. (1) in the frame of the moving pulse. With the standard substitution \( \eta = t - z/u \), where the quantity \( \eta \) is the local time, which is the shift of time origin in point of pulse maximum. Then Eq. (1) is transformed into \[ \frac{\partial A(z, \eta)}{\partial z} - i \frac{k_2}{2} \frac{\partial^2 A(z, \eta)}{\partial \eta^2} = 0. \] (2)

Using Fourier transform,

\[ A(\eta) = \int_{-\infty}^{\infty} \hat{A}(\Omega) e^{i\Omega \eta} d\Omega, \]

we obtain the following equation for the fundamental spectral amplitude

\[ \frac{\partial \hat{A}}{\partial z} + i \frac{k_2 \Omega^2}{2} \hat{A} = 0, \] (3)

where \( \Omega \) is the deviation from the central frequency of the output wave \( \Omega = \omega - 2\omega_0 \) for the SHG process and \( \Omega = \omega - \omega_0 \) for the XPW generation process.

The solving of the equation as a function of \( z \) is

\[ \hat{A} = \hat{A}(z, \Omega) = \hat{A}_0(\Omega) e^{-\frac{k_2 \Omega^2 z}{2}}. \] (4)

This is the solution for the evolution of the spectrum when it propagates through dispersive media. The input wave spectrum could have linear and quadratic chirp. Then the result for the output spectral amplitude will have the following form:

\[ \hat{A}(z, \Omega) = \hat{A}_0 e^{-\frac{\Omega^2}{2\sigma^2}} e^{i \phi_2 \frac{\Omega^2 z}{2}} + i \phi_3 \frac{\Omega^3 z}{6} - \frac{k_2 \Omega^2 z}{2} \]

(5)

where \( \sigma \) is the intensity spectral half width at 1/e level, \( \phi_2 \) and \( \phi_3 \) are the second and the third order spectral phases. From Eq. (5) it is seen that if the input wave has only linear chirp (only second order phase term is not zero) the dispersion of the media \( k_2 z \) can compensate the linear chirp at distance \( z = \phi_2/k_2 \) [9].

In what follows we will consider relatively short nonlinear media, so the term \( \exp(-i \frac{k_2 \Omega^2 z}{2}) \) will be neglected in Eq. (5) and the fundamental spectrum will be considered not \( z \) dependent.

2.2 Spectral Approach for SHG Process

SHG is a nonlinear process where the output wave has two times bigger frequency. The propagation equation for the second harmonic \( B_{\text{SHG}} \) generated by a fundamental wave with spectral width \( \sigma \) and spectral phase \( \phi(\omega) = \)
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\[ \frac{1}{2} \phi_2 \Omega^2 + \frac{1}{6} \phi_3 \Omega^3 \] (these are the first two terms of the Taylor series) in approximation of non-depletion has the following form:

\[ \frac{\partial B_{\text{SHG}}(z, \eta)}{\partial z} - \frac{i k_2}{2} \frac{\partial^2 B_{\text{SHG}}(z, \eta)}{\partial \eta^2} = -i \gamma A(z, \eta) A(z, \eta), \quad (6) \]

where \( \gamma \) is the second order nonlinear coupling coefficient. After Fourier transform

\[ B_{\text{SHG}}(z, \eta) = \int_{-\infty}^{\infty} \hat{B}_{\text{SHG}}(z, \Omega) e^{i \Omega \eta} d\Omega, \]

\[ A(z, \eta) = \int_{-\infty}^{\infty} \hat{A}(z, \omega_1) e^{i \omega_1 \eta} d\omega_1 \]

one obtains

\[ \frac{\partial \hat{B}_{\text{SHG}}(z, \Omega)}{\partial z} + i \frac{k_2}{2} \hat{B}_{\text{SHG}}(z, \Omega) \Omega^2 = -i \gamma K_{\text{SHG}}(z, \Omega), \quad (7) \]

where

\[ K_{\text{SHG}}(z, \Omega) = \int_{-\infty}^{\infty} \hat{A}(z, \omega_1) \hat{A}(z, \Omega - \omega_1) d\omega_1 \]

is the autocorrelation function of the fundamental spectral field. Neglecting second order dispersion of the nonlinear medium we have

\[ \hat{B}_{\text{SHG}}(z, \Omega) = -i \gamma z K_{\text{SHG}}(\Omega). \quad (8) \]

Let us obtain correlation function when the complex electric field consists of Gaussian spectral amplitude and linear and quadratic frequency chirp, \textit{i.e.}

\[ \hat{A}(z, \omega_1) = \hat{A}_0 e^{-\frac{\omega_1^2}{2 \sigma^2}} \exp \left[ i \left( \frac{\phi_2}{2} (\omega_1)^2 + \frac{\phi_3}{6} (\omega_1)^3 \right) \right] \quad \text{and} \]

\[ \hat{A}(z, \Omega - \omega_1) = \hat{A}_0 e^{-\frac{(\Omega - \omega_1)^2}{2 \sigma^2}} \exp \left[ i \left( \frac{\phi_2}{2} (\Omega - \omega_1)^2 + \frac{\phi_3}{6} (\Omega - \omega_1)^3 \right) \right] \]

yields

\[ K_{\text{SHG}}(\Omega) = \frac{\hat{A}_0^2 \sqrt{\pi} e^{-\frac{\Omega^2}{4 \sigma^2}} \exp \left[ i \left( \frac{\phi_2}{4} (\Omega)^2 + \frac{\phi_3}{24} (\Omega)^3 \right) \right]}{\sqrt{\sigma^{-4} + (\phi_2 + \frac{1}{2} \Omega \phi_3)^2}}. \quad (9) \]

We obtain analytical expressions for spectral intensity \( I_{\text{SHG}} \) of the pulses with doubled frequency

\[ I_{\text{SHG}}(\Omega) = \frac{\varepsilon_0 c \eta}{2} \frac{\gamma^2 \sigma^2 \varepsilon_0^2 \hat{A}_0^4 e^{-\frac{\Omega^2}{2 \sigma^2}}}{\sqrt{1 + \sigma^2 (\phi_2 + \frac{1}{2} \Omega \phi_3)^2}} \quad (10) \]
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From comparing the exponential terms in Eq. (9) and Eq. (5) it is clearly seen that the second order spectral phase of the output SH wave is twice smaller than the second order spectral phase of the fundamental beam and that third order spectral phase of the output SH wave is four times smaller than the input third order spectral phase. This conclusion is important since it shows that the nonlinear process reduces the spectral phase distortions. From Eq. (10) we conclude that the presence of second order phase (linear chirp) does not influence the width and shape of SH output spectrum. The nonzero \( \phi_2 \) just reduces the SH intensity amplitude. Also from Eq. (10) it is easy to obtain the quantity proportional to the energy of the SH pulses by simple integration of this expression using the next definition:

\[
W_{\text{SHG}} \propto \int_{-\infty}^{\infty} I_{\text{SHG}}(\Omega) d\Omega.
\]  

(11)

Indeed, for example for the case of \( \phi_3 = 0 \), we have

\[
W_{\text{SHG}} \propto \frac{2\gamma^2\sigma_3^4A_0^4\sqrt{2\pi\sigma}}{\sqrt{1 + (\phi_2)^2\sigma^4}}.
\]  

(12)

This expression directly shows that the presence of linear chirp in the fundamental pulse reduces the SH energy and the efficiency by factor \( 1/\sqrt{1 + (\phi_2)^2\sigma^4} \).

The presented numerical results in this paper are obtained with input pulse with FWHM duration 30 fs that corresponds to spectral width \( \sigma \) on \( 1/e \) intensity level 56 THz. For this and longer pulse durations and nonlinear media with

![Figure 1. SHG spectra for input pulse with quadratic chirp only](image_url)
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length 1.2 mm neglecting the second order dispersion of index of refraction is acceptable. If we consider shorter pulses or longer nonlinear media then we have to consider the refractive index changes into the frames of spectral shape and the model won’t be accurate.

Using Eq. (10) we obtain the SHG intensity spectra normalized to its value without input spectral phase terms. They are shown in Figure 1 and Figure 2. The spectra are shown only for positive values of the phases due to the fact that \( I_{\text{SHG}}(\Omega, \phi_2) = I_{\text{SHG}}(-\Omega, -\phi_2) |_{\phi_3=\text{const}} \) and \( I_{\text{SHG}}(\Omega, \phi_3) = I_{\text{SHG}}(-\Omega, -\phi_3) |_{\phi_2=\text{const}} \) which we can see from Eq. (10), i.e. the spectra for

![Figure 2. SHG spectra for different values of quadratic and cubic phases: (top) for \( \phi_2 = 1000 \, \text{fs}^{-2} \); (bottom) for \( \phi_2 = 2000 \, \text{fs}^{-2} \).](image)
negative phases are symmetrical to the shown ones. We note that when it has only one of both phases spectrum it is Gaussian and only its width is changed. When it has nonzero terms of both phases the spectrum degenerates. With increasing of the third order phase the spectral broad reduces and fastly increases spectral maximum which is in low frequency domain (high $\lambda$). Increasing of quadratic phase helps increasing of the maximum but weakly increases spectrum width.

Dependences of SHG energy on quadratic and cubic spectral phases normalized to the value of the energy of Fourier transformed limited are shown in Figure 3. At constant cubic phase the energy is Lorentzian-like function of the quadratic

![Figure 3. SHG energy for different values of quadratic and cubic phases: (top) for fixed values of $\phi_3$; (bottom) for fixed values of $\phi_2$.](image-url)
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phase. When the quadratic phase is constant the energy becomes a more complex function of the cubic phase with two symmetrical maxima. Numerical simulations are obtained by solving Eq. (11) and when the spectral phase values are negative, the curves are completely the same due to the fact that “the areas” under the curves of the spectra are equal.

Figure 4 shows the behavior of the spectral width. The definition of the spectral width we use here is

$$\Delta \Omega = \frac{\int_{-\infty}^{\infty} I(\Omega) d\Omega}{I_{\text{max}}(\Omega)}$$

(13)

Figure 4. SHG spectral width for different values of quadratic and cubic phases: (top) for fixed values of $\phi_3$; (bottom) for fixed values of $\phi_2$. 95
The curves in Figure 4 are normalized to its value when its spectral phases of second and higher order are zero. In absence of cubic phase the spectral width is constant. This can be proved analytically. Applying definition (13) with respect to intensity Eq. (10) we obtain that second order spectral phase does not influence the spectral width that is constant $\Delta \Omega = \sqrt{2\pi} \sigma$ exactly $\sqrt{2}$ broader than the width of the fundamental spectrum.

If there is no quadratic phase, the dependency is Lorentzian, and in its presence the curves have two symmetrical maxima. Figure 5 shows the phase behavior.

Figure 5. SHG barycenter shift for different values of input quadratic and cubic phases: (top) for fixed values of $\phi_3$; (bottom) for fixed values of $\phi_2$. 

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of the position of the barycenter (with respect to the central frequency for the process) normalized to the width of spectrum without any phase terms. The definition of the barycenter we use here is

\[ B_{C} = \frac{\int_{-\infty}^{\infty} \Omega I(\Omega) d\Omega}{\int_{-\infty}^{\infty} I(\Omega) d\Omega}. \]  

(14)

The position of the barycenter is indication of the spectral shift of the output spectral shape.

Figure 6. 3D (top) and contour (bottom) dependences of SH pulse energy on input second and third order phases.
From Eq. (14) we can see that it is an odd function due to the energy, it is even function with spectral symmetry. From the definition of the quantity we could note that it is analogous to the mass center of mechanical system, i.e. it depends strongly on the shape of spectrum. In the presence only of one of both spectral phases there is no barycenter shift. In this case one can observe only change of the spectral width, but not the shift. The barycenter shift in this figure is normalized to the value of the spectral width of transform limited input pulse.

In Figures 6-8 three dimensional characteristics of the investigated quantities are shown.
2.3 Spectral Approach for XPW Process

In a similar way as we introduced the effect of second harmonic generation we would consider the effect of cross polarized wave generation in cubic crystals. The effect describes the generation of a new wave polarized perpendicularly to the input polarization. We will again use slowly varying envelope approximation $\frac{\partial^2 A}{\partial z^2} \ll k\frac{\partial A}{\partial z}$ with neglect of linear and nonlinear absorption, the self-phase modulation, cross-phase modulation, four-wave mixing processes and also the depletion of the fundamental pulse. In the frame of the coordi-
nate system formed by the input and output polarization we obtain the following plane wave equation for the field $B_{X PW}$ propagating along crystallographic axis $z$ [10]:

$$
\frac{\partial B_{X PW}(z,t)}{\partial z} + \frac{1}{u} \frac{\partial B_{X PW}(z,t)}{\partial t} - \frac{i k_2}{2} \frac{\partial^2 B_{X PW}(z,t)}{\partial t^2} = i \gamma_\perp A(z,t) A^*(z,t) A(z,t), \tag{15}
$$

where $\gamma_\perp = -\gamma_0 \frac{\sigma_x}{4} \sin(4\beta)$, $\gamma_0 = \frac{6\pi}{8\lambda_0} \chi^{(3)}_{xxx}$ and $\sigma_x$ is the anisotropy of the $\chi^{(3)}$ tensor. $\beta$ is the angle between the input polarization and the crystallographic axis $x$. It is convenient to rewrite (15) in the frame of the moving pulse.

With the standard substitution $\eta = t - z/u$, it becomes

$$
\frac{\partial B_{X PW}(z,\eta)}{\partial z} - \frac{i k_2}{2} \frac{\partial^2 B_{X PW}(z,\eta)}{\partial \eta^2} = i \gamma_\perp A(z,\eta) A^*(z,\eta) A(z,\eta). \tag{16}
$$

Using Fourier transform $B_{X PW}(z,\eta) = \int_{-\infty}^{\infty} \hat{B}_{X PW}(z,\Omega) e^{i\Omega \eta} d\Omega$ and applying the reverse convolution theorem, we obtain the following equation for the complex spectral amplitude of the XPW pulse:

$$
\frac{\partial \hat{B}_{X PW}(z,\Omega)}{\partial z} + i \frac{k_2}{2} \hat{B}_{X PW}(z,\Omega) \Omega^2 = -i \gamma_\perp K_{X PW}(z,\Omega) \tag{17}
$$

with the third order spectral autocorrelation function defined as

$$
K_{X PW}(z,\Omega) = \int_{-\infty}^{\infty} \tilde{A}(z,\omega_1) \tilde{A}(z,\omega_2) \tilde{A}^*(z,\omega_1 + \omega_2 - \Omega) d\omega_1 d\omega_2. \tag{18}
$$

The frequencies $\omega_1, \omega_2, \omega_3 = \omega_1 + \omega_2 - \Omega$ are deviations from the same central frequency for all four interacting waves: XPW and the three input waves, respectively. Since we here consider short pulses with broad spectrum we have used in (18) that four wave mixing between all spectral components takes place and consequently $\Omega = \omega_1 + \omega_2 - \omega_3$. Let us assume non-depletion regime and if we consider the fundamental field in form of (5), then XPW output spectral amplitude will have the form:

$$
\hat{B}_{X PW}(L,\Omega) = -i \gamma_\perp e^{-i \frac{k_2}{2} \Omega^2} \int_{0}^{L} K_{X PW}(z,\Omega) e^{i \frac{k_2}{2} \Omega^2} dz. \tag{19}
$$

As we have discussed before, for short enough nonlinear media and not so short pulses, the second order dispersion $k_2$ can be neglected and the fundamental amplitude will be not $z$ dependent. Then the complex spectral XPW output amplitude will be

$$
\hat{B}_{X PW}(\Omega) = -i \gamma_\perp K_{X PW}(\Omega) L. \tag{20}
$$
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Eq. (20) can be solved analytically only for the specific case of single $\phi_2 \neq 0$ and all other phases equal to zero. In this case the XPW spectral amplitude can be expressed in analytical form as follows:

$$\hat{B}_{XPW}(\Omega) = \frac{-i2\pi\gamma L^2 A_0^3 L}{\sqrt{(\phi_2 \sigma^2 - 3i)(\phi_2 \sigma^4 + i)}} \times \exp \left[ -\frac{\Omega^2}{6\sigma^2} + \frac{i\phi_2 \Omega^2}{9} \left( 1 + \frac{\phi_2^2 \sigma^4}{1 + \frac{\phi_2^2 \sigma^4}{9}} \right) \right]. \quad (21)$$

For transform limited (unchirped pulse) the XPW spectral amplitude will be

$$\hat{B}_{XPW}(L, \Omega) = \frac{-i2\pi\gamma L^2 A_0^3 L}{\sqrt{3}} \exp[-\Omega^2/(6\sigma^2)]. \quad (22)$$

So the output spectrum will be $\sqrt{3}$ times broader and respectively the pulse will be $\sqrt{3}$ shorter.

From Eq. (21) the spectral intensity of the XPW can be found

$$I_{XPW}(L, \Omega) = \frac{\varepsilon_0 cn^2}{2} \frac{(2\pi\gamma L^2 A_0^3 L)^2}{(\phi_2^2 \sigma^4 + 9)(\phi_2^2 \sigma^4 + 1)} \times \exp \left[ -\frac{\Omega^2}{3\sigma^2} \left( 1 + \frac{\phi_2^2 \sigma^4}{1 + \frac{\phi_2^2 \sigma^4}{9}} \right) \right]. \quad (23)$$

The output spectral width of the XPW will be different from the input width by

![Figure 9. XPW spectra for zero second order phase and fixed values of third order phase](image)

Figure 9. XPW spectra for zero second order phase and fixed values of third order phase
the factor that depends on the normalized input chirp. Indeed using the definition (13) we obtain

$$\sigma_{\text{XPW}} = \sqrt{\frac{3(1 + \phi_2^2 \sigma^4/9)}{(1 + \phi_2^2 \sigma^4)}}. \quad (24)$$

As seen from (24) depending on the input chirp the output width is changing from maximum $\sigma \sqrt{3}$ for very small input quadratic chirps to minimum value $\sigma / \sqrt{3}$ for very big input quadratic phase.

Analyzing (21) we find out how the output XPW second order phase depends on
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Figure 11. XPW pulse energy vs input second and third order phases: (top) for fixed values of $\phi_3$; (bottom) for fixed values of $\phi_2$.

the input second order phase in case of all higher order phases are zero. Indeed

$$\phi_{2,\text{out}} = \phi_2 \frac{(1 + \phi_2^2 \sigma^4)}{9(1 + \phi_2^2 \sigma^4/9)}. \quad (25)$$

It is clear from (25) the XPW process can suppress not very big chirps, when $\phi_2 \sigma^2 \ll 1$. Depending on the input chirp the output second order phase is changing from $\phi_2/9$ minimum value for very small input chirp to maximum value $\phi_2$ for very big input chirps.

Let us find the quantity proportional to the energy of the pulses by integration of
As can be seen from (26) the input linear chirp reduces the XPW efficiency. Experimental results confirm this. Note that such dependences have been reported in [11].

For the case when both $\phi_2 \neq 0$ and $\phi_3 \neq 0$ the Equation (19) is solved numerically.
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Figure 13. XPW barycenter shift vs input second and third order phases: (top) for fixed values of $\phi_3$; (bottom) for fixed values of $\phi_2$.

In Figure 9 and Figure 10 are shown XPW spectra normalized to the value of the intensity without spectral phase terms. Due to similar considerations as we have done for SHG, the dependences shown are only for positive terms of the spectral phase. From these figures we see that it is enough one of the two phases to be zero, in order the output XPW spectrum to be Gaussian or close to Gaussian. With increase of both phases the spectral shapes become asymmetric with high frequency slope steeper than the other one, and close to the steep slope small spectral pick appears.
If the phases are negative the spectral shape is reversed putting the steeper slope from the lower frequency part. It is curious to note that spectral maximum is always at almost zero frequency shift, i.e. at frequency equal to central frequency of the input signal.

With numerical simulations we obtain dependences (shown in Figure 11) of the XPW pulse energy on the second and third order phases. The curves are normalized to the magnitude of the energy of Fourier transformed limited pulse. From plots we see that they qualitatively repeat the obtained for SHG.
Let us consider numerical calculations for the spectral width as a function of the second and third order phases and normalized to the width of spectrum without phase terms. They are shown in Figure 12. They are similar by shape with those curves we have considered for the energy. The curves of spectral width are with more sloping slopes which is due to the shift of the maximum of the intensity.

The results for the barycenter shift as a function of the quadratic phase at constant cubic phase and as a function of the cubic phase at constant quadratic phase are shown in Figure 13. They again are qualitatively similar to the obtained for SHG. If the spectral phases are zero then the value of the barycenter shift is
zero as well. The barycenter shift in this figure is normalized to the value of the spectral width of transform limited input pulse. With the increasing of the phases barycenter shift is reaching a maximum followed by reduction.

Relying on a full model we present in Figures 14-15 three dimensional dependences for the energy, spectral width and barycenter shift as a function of second and third order phases.
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3 Conclusions

We have presented a study of the influence of the input phase constants on spectral properties of the output of nonlinear optical processes second harmonic generation and cross polarized wave generation. The results allow proper design of femtosecond systems with nonlinear optical converters of the frequency or laser beam parameter. We show here that nonlinear optical converters reduce the input phase constants. By controlling the input phases blue or red shift of the generated second harmonic wave can be achieved.

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