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Spectral broadening and pulse duration reduction during cross-polarized wave generation: influence of the quadratic spectral phase

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ABSTRACT Cross-polarized wave (XPW) generation is used for the contrast improvement of ultra-intense femtosecond laser pulses in a double CPA configuration. We present theoretical and experimental evidence that the XPW output spectrum depends in a predictable way on the input chirp. Therefore, a chirp controlled pulse can experience a pulse duration shortening up to a factor of $\sqrt{3}$, and an initial amount of chirp that leads to the exact preservation of the spectral width of a given pulse can be predicted.

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1 Introduction

Cross-polarized wave (XPW) generation is a degenerated four-wave mixing process occurring in a nonlinear medium whose third order nonlinear susceptibility is anisotropic [1–3]. This process allows the conversion of part of an input linearly polarized wave into an orthogonally polarized wave. The relevance of this mechanism on the design of nonlinear filters devoted to contrast enhancement of ultra-intense femtosecond laser pulses has formerly been demonstrated [4–6]. It has been established that filters based on XPW generation allow improvement of pulse contrast by several orders of magnitude without introducing any spatial or spectral distortions. Considering the crucial importance of the temporal contrast ratio for the development of petawatt class laser chains, we have also shown in previous work the advantages of the XPW nonlinear filter in terms of transmission efficiency and contrast improvement [7, 8].

Here, we theoretically and experimentally investigate the temporal and spectral characteristics (intensity and phase) of the generated cross-polarized wave according to the input pulse duration and chirp. Indeed, a way to produce pulses with both high contrast and high peak power is to achieve a double CPA system including a XPW filter [9]. Knowing the temporal and spectral properties of the cleaned pulse is consequently of prime importance to the design of the second CPA setup.

Furthermore, an interesting point is that a XPW generated signal presents, in a first approximation, a cubic dependence on intensity of the input signal. Therefore, if we assume that pulses exhibit a usual temporal Gaussian shape, the duration of the XPW signal should be shorter than the input one and its spectrum should be broadened. This process has experimentally been observed in [5] and is a very attractive method for producing ultrashort pulses. In that case, the nonlinear filter allows for the compensation of eventual gain spectral narrowing in the first CPA and for the production of shorter pulses with high temporal contrast. This paper demonstrates the conditions on the input pulse to verify this hypothesis. We have particularly investigated the influence of the initial residual spectral phase (chirp), which is easily introduced by optical elements preceding the nonlinear medium. We will show that this value has a crucial impact on the duration of the XPW signal. We have shortened down to 14 fs a 23 fs pulse with XPW by suppressing its initial chirp. Moreover, we demonstrate that for such short pulses an excessive amount of chirp leads to spectrum narrowing and pulse duration broadening, preventing us from using XPW generation for contrast filtering in the uncompressed picosecond regime.

2 Theoretical considerations

2.1 *Hypothesis*

We consider spectral and associated temporal Gaussian shapes for input and XPW pulses. We also consider linear chirp in this paper; the influence of the coefficients of spectral phase with order higher than two is neglected in our investigation.

The effective full-width at half maximum (FWHM) duration of the initial

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pulse is denoted as Δt and Δt_{XPW} for the XPW generated pulse. Their Fourier transform limited FWHM duration is Δt_0 and $\Delta t_{0\mathrm{XPW}}$. The second order spectral phase coefficient, or chirp, is denoted as φ (fs^2) for the incoming laser pulse and φ_{XPW} for the XPW generated pulse. These parameters are linked by the well-known formula, valid for a Gaussian chirped pulse [10]:

$$\Delta t = \Delta t_0 \sqrt{1 + 16\ln^2(2)\frac{\varphi^2}{\Delta t_0^4}}\,. \tag{1}$$

We aim to determine the values of Δt_{XPW} and φ_{XPW} and their dependence with initial pulse characteristics. The Fourier transform limited duration $\Delta t_{0\mathrm{XPW}}$ and associated spectral width of the XPW output will then be known.

The expression of the electric field spectrum corresponding to the initial chirped pulse with carrier frequency ω_0 can be written as:

$$E(\omega) = E_0 \exp\left(-\frac{(\omega-\omega_0)^2}{4\Gamma_0}\right) \times \exp\left(j\frac{\varphi}{2}(\omega-\omega_0)^2\right)\,. \tag{2}$$

The term Γ_0 is related to the input unchirped pulse duration Δt_0 by $\Gamma_0 = \frac{2\ln(2)}{\Delta t_0^2}$.

In the temporal domain, the input field $E(t)$ is obtained by the inverse Fourier transformation of (2):

$$E(t) = \frac{1}{2\pi} E_0 \exp(-\Gamma' t^2)\exp(-j\omega_0 t)\,. \tag{3}$$

We introduce in (3) a complex parameter $\Gamma' = (1/\Gamma_0 - 2j\varphi)^{-1}$. The expression for the electric field $E(t)$ then takes the form:

$$E(t) = \frac{1}{2\pi} E_0 \exp(-(\Gamma + j\varphi)t^2) \times \exp(-j\omega_0 t) \tag{4}$$

where $\Gamma = \frac{\Gamma_0}{1+4\varphi^2\Gamma_0^2}$ and $\varphi = \frac{2\Gamma_0\varphi}{1+4\varphi^2\Gamma_0^2}$. The second order temporal phase of the pulse is given by φ. Similarly to Γ_0, the Γ parameter is linked to the effective duration Δt of the input chirped pulse: $\Gamma = \frac{2\ln(2)}{\Delta t^2}$. A simple relationship between φ and Γ, which is true for a Gaussian chirped pulse, can then be extracted:

$$\varphi = 2\varphi\Gamma_0\Gamma\,. \tag{5}$$

2.2 *Spectral and temporal characteristics of the XPW generated pulse*

We can now determine the characteristics of the XPW pulse knowing the input pulse characteristics. XPW generation is a frequency degenerated third order nonlinear effect. Consequently, for a monochromatic wave, the related nonlinear polarization can be expressed as:

$$P^{(3)}(\omega) \propto \chi^{(3)} E^*(\omega)E(\omega)E(\omega)\,. \tag{6}$$

As ultra-short pulses are not monochromatic, various frequencies included in the spectral width of the pulse contribute to the four wave mixing process. In this case, the nonlinear polarization has to be written as a convolution product. This convolution takes into account all possible frequency combinations for each spectral component. Equation (6) becomes:

$$P^{(3)}(\omega) \propto \chi^{(3)} E^*(\omega) \otimes E(\omega) \otimes E(\omega) \tag{7}$$

then

$$P^{(3)}(t) \propto \widetilde{\chi}^{(3)} E^*(t)E(t)E(t)\,. \tag{8}$$

As the generated field is proportional to $P^{(3)}$ and using (4), we deduce an expression for $E_{\mathrm{XPW}}(t)$ at the frequency ω_0:

$$E_{\mathrm{XPW}}(t) = E_0' \exp(-\Gamma_{\mathrm{XPW}} t^2) \times \exp(-j\varphi_{\mathrm{XPW}} t^2 - j\omega_0 t)\,. \tag{9}$$

The new parameters Γ_{XPW} and φ_{XPW} are expressed as follows:

$$\Gamma_{\mathrm{XPW}} = 3\Gamma \tag{10}$$

$$\varphi_{\mathrm{XPW}} = \varphi\,. \tag{11}$$

Similarly to Γ_0, Γ_{XPW} is linked to the effective duration of the XPW pulse Δt_{XPW} while φ_{XPW} is the temporal phase of the generated pulse. Equation (11) demonstrates that the temporal shape of the pulse is unchanged during the process.

Considering the definition of Γ and (10), and also (5) and (11) we obtain:

$$2\Gamma_{0\mathrm{XPW}}\Gamma_{\mathrm{XPW}}\varphi_{\mathrm{XPW}} = 2\Gamma_0\Gamma\varphi\,. \tag{12}$$

Knowing that $\Gamma_{0\mathrm{XPW}}$ is related to the Fourier transform limited duration of the generated pulse Δt_{XPW} by $\Gamma_{0\mathrm{XPW}} = \frac{2\ln(2)}{\Delta t_{0\mathrm{XPW}}^2}$, we finally obtain expressions for Δt_{XPW} and φ_{XPW}:

$$\Delta t_{\mathrm{XPW}} = \frac{\Delta t}{\sqrt{3}} \tag{13}$$

$$\varphi_{\mathrm{XPW}} = \frac{\varphi}{3}\frac{\Delta t_{\mathrm{XPW}}^2}{\Delta t_0^2}\,. \tag{14}$$

The effective duration of the XPW pulse is then fully known and is $\sqrt{3}$ shorter than the effective duration of the initial pulse. Using (1) associated to the generated pulse, (13) and (14) lead to the expression of the Fourier transform limited XPW pulse duration:

$$\Delta t_{0\mathrm{XPW}} = \frac{\Delta t_{\mathrm{XPW}}}{\sqrt{1 + 16\ln^2(2)\frac{\varphi^2}{9\Delta t_0^4}}}\,. \tag{15}$$

As Δt_{XPW} and Δt are known, $\Delta t_{0\mathrm{XPW}}$ and φ_{XPW} are deducible from (15):

$$\Delta t_{0\mathrm{XPW}} = \frac{\Delta t_0}{\sqrt{3}} Z(\Delta t_0, \varphi) \tag{16}$$

$$\varphi_{\mathrm{XPW}} = \frac{\varphi}{9} Z^2(\Delta t_0, \varphi)\,, \tag{17}$$

with

$$Z(\Delta t_0, \varphi) = \frac{\sqrt{1 + 16\ln^2(2)\frac{\varphi^2}{\Delta t_0^4}}}{\sqrt{1 + 16\ln^2(2)\frac{\varphi^2}{9\Delta t_0^4}}}\,. \tag{18}$$

Expression (16) shows that for a negligible chirp the pulse can be shortened and we cover this important property of XPW in the next section. We can notice also from (17) that for a moderate input chirp, i.e., $\varphi \ll \Delta t_0^2/3$, the output chirp is dramatically reduced. While for larger chirp, i.e., $\varphi \gg \Delta t_0^2/3$, the output chirp is kept the same as the input one.

2.3 *XPW generation and spectral broadening*

Equation (16) points out the potential shortening of FT limited duration of the pulse during the conversion process. The $\sqrt{3}$ reduction coefficient is corrected by the $Z(\Delta t_0, \varphi)$ term, defined by (18), which strongly depends on Δt_0 and φ, the initial pulse duration and chirp of the input pulse.

The evolution of Z with the initial chirp value is plotted in Fig. 1a for

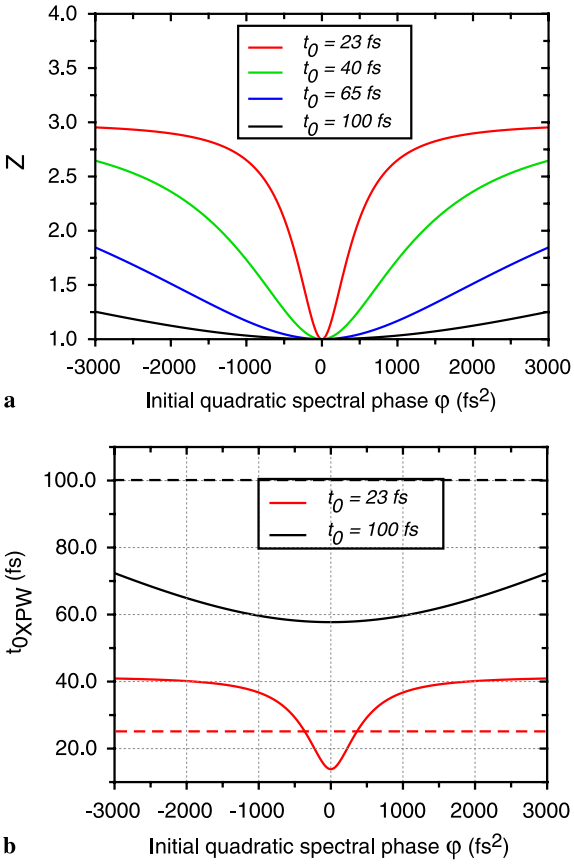


FIGURE 1 (**a**) Dependence of Z with the initial quadratic spectral phase value for different initial pulse durations. (**b**) Theoretical dependence of the Fourier transform limited duration of the XPW pulse (*full lines*) with the initial quadratic spectral phase value for different initial pulse durations (*dashed lines*)

different values of Δt_0. This calculation will lead to a theoretical estimation of the XPW pulse Fourier transform limited duration, as illustrated in Fig. 1b.

It appears that the evolution of Z with the chirp is dependent on the initial Fourier transform limited pulse duration. For relatively long pulses, 100 fs or more, Z value remains close to 1 whatever the chirp value is. Consequently, thanks to (16) we can deduce that the XPW pulse FT limited duration will be reduced by a factor close to $\sqrt{3}$ for any input long pulse, i.e., $\sim$ 100 fs, with any experimental input chirp as long at it keeps the pulse duration below 1 ps. This is understood as second order phase has relatively low influence on such long pulse alike second order phase added by optical elements. This phenomenon is visible in Fig. 1b and has been previously measured experimentally [5]. This makes the XPW process an easy way to shorten long femtosecond pulses. The damage threshold is limiting XPW generation below the 1 ps pulse duration, and above this limit, thermal damage is induced by the laser in the crystal.

With shorter pulses the chirp will have a stronger influence on the Z value. Consequently, the pulse shortening will depend on the input pulse parameters. In every case, the initial quadratic spectral phase has to be very close to zero during the interaction, in order to allow for pulse duration reduction and the corresponding spectral broadening. The best reduction (giving a $\sqrt{3}$ factor reduction) is always obtained when the input pulse is perfectly compressed during the nonlinear process. The compensation of all optical elements preceding the filter is of prime importance.

Furthermore, for short pulses, about 40 fs or less, we can extract more information from Fig. 1. The shortening of the pulse by a $\sqrt{3}$ factor is always possible if $\varphi = 0$ fs^2, but for significant chirp value, depending on the value of Δt_0, the model predicts that the Fourier transform pulse duration after XPW (Δt_{0XPW}) can become superior to the input Fourier transform pulse duration (Δt_0). This means that the spectrum can be narrowed during the process. Indeed, if the initial spectrum is large, second order spectral phase can lead to an important linear temporal spreading of spectral frequencies. Extreme frequencies are then included in the temporal wings of the pulse, less intense than the peak. These frequencies will not be efficiently converted into the XPW signal and this phenomenon will introduce a spectral narrowing. Figure 1b illustrates the case of an input pulse with a FT limited duration of 23 fs. For such a pulse, the XPW pulse will be longer than the initial one, if $|\varphi| > 400$ fs^2. This observation points out again the necessity to control the initial quadratic phase for short pulses. This property of the XPW process prevents the use of this filter for contrast filtering on a stretched pulse; XPW contrast filtering has to be performed in the double CPA configuration. We can define a critical initial chirp value, φ_{CR}, for which the XPW pulse duration will be identical to the input pulse. The φ_{CR} expression is deduced from (16) and (18):

$$\varphi_{CR} = \frac{\sqrt{3}}{\ln(2)} \left(\frac{\Delta t_0}{2} \right)^2 , \qquad (19)$$

and φ_{CR} quadratic dependence with Δt_0 is plotted in Fig. 2. For ultra-short pulses, as sub-10 fs pulses, a dispersion

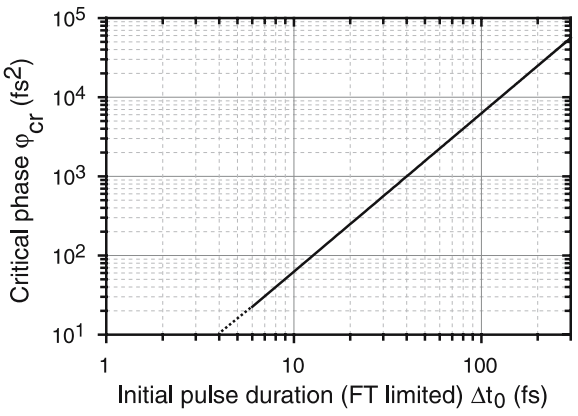


FIGURE 2 Theoretical critical quadratic spectral phase value (chirp) as a function of initial pulse duration

of a few tens of femtoseconds squared can be enough to narrow the spectrum. For such pulses, which often suffer from a poor coherent contrast [11–14] and so could be candidates for XPW filtering, the accurate control of second order spectral phase is mandatory and can be exerted by an acousto-optic programmable dispersive filter (AOPDF) or chirped mirrors [15]. In that case, the dispersion of the nonlinear crystal itself has to be taken into account. An adequate medium for XPW generation is barium fluoride crystal (BaF_2, cubic symmetry) [3, 4]. The quadratic spectral phase introduced by this crystal is about $380\,\mathrm{fs^2cm^{-1}}$, which is low and comparable to the one of fused silica. This value is obtained by simple derivation of the formula giving the refractive index as a function of wavelength [16–18]. In a first approximation, to obtain a XPW process efficiency of at least 10%, the minimum length of the BaF_2 crystal must be 1 mm, and introduces during the process a dispersion in the range of $40\,\mathrm{fs^2}$. According to Fig. 2, this is a critical phase value for a 8 fs (FT limited) input pulse. For shorter pulses (i.e., 6 fs), a less dispersive medium also suitable for XPW generation such as CaF_2 ($250\,\mathrm{fs^2cm^{-1}}$ [18]) could be used.

XPW generation in cubic crystals is therefore adapted to filter sub-10 fs pulses. During the nonlinear effect, the spectrum should not be narrowed, but we cannot quantify an eventual broadening with the presented model, since sub-10 fs pulses do not exhibit Gaussian nor smooth spectra.

For moderate XPW conversion efficiency and a Gaussian input pulse, the spectral width of the generated XPW pulse is related to the input spectral width by the expression:

$$\Delta\nu_{\mathrm{XPW}} = \Delta\nu_0 \frac{\sqrt{3}}{Z(\Delta t_0, \varphi)}\,. \tag{20}$$

We use this expression to compare the above described model with experiments.

3 Experiments

In order to validate the theoretical model described in this paper, experiments have been performed with two different femtosecond lasers. The XPW signal is characterized in both spectral and temporal fields.

3.1 *Experimental setups*

The first experiments have been conducted with a chirped pulse amplification laser, emitting pulses with duration of 65 fs (FT limited), with a maximum energy of 70 μJ at 1 kHz repetition rate. The Gaussian spectrum of the pulses measures 15 nm (FWHM) and is centered at 820 nm. Positive or negative second order spectral phase (φ, $\mathrm{fs^2}$) is added by changing the compressor adjustment. The reached spectral phase range is $\pm 3000\,\mathrm{fs^2}$ and the maximum chirped pulse duration is then 140 fs.

The input pulse is seeded in the XPW filter, which includes crossed polarizers and a 700 mm focalization lens. The nonlinear medium is a 2 mm long BaF_2 crystal placed at the focus. The quadratic spectral phase introduced by the BaF_2 crystal is neglected for this pulse duration. This XPW efficiency value is 5%, which is low enough to assure an unsaturated XPW generation without phase mismatch due to SPM [8]. This efficiency value for one crystal corresponds to the range where the XPW signal presents a pure cubic dependence with the input pulse intensity [3].

We have also used a chirped pulse amplification laser, emitting pulses with energy of 250 μJ at 10 Hz repetition rate. The Gaussian spectrum of the pulses measures 49 nm (FWHM) and is centered at 800 nm. At the output of the laser, the measurement of the pulse duration gives 23 fs (Fig. 5), which is not the FT limited duration associated with the experimental spectrum. Indeed, the remaining spectral phase is not flat and high orders are not totally compensated.

The second XPW filter is composed of crossed polarizers and a 1500 mm focalization lens. Two 2 mm long BaF_2 crystals separated by a distance of 250 mm are used to improve the efficiency of the overall XPW process [7, 8]. The double crystal setup is used to overcome the saturation issue of the process. The saturation ends the cubic dependence on intensity of the XPW signal and occurs above 10% efficiency for a single crystal scheme. This saturation process is due to a nonoptimal phase relation between the incoming and the XPW generated pulses due to SPM. Therefore, the use of two thin crystals is not just an increase of the nonlinear medium length, but takes advantage of the Kerr lens effect refocusing differently the input and XPW pulse and the subsequent Gouy phase difference between the two pulses, to achieve an appropriate phase relation between main and XPW waves in the second crystal. In these conditions, the cubic dependence is prolonged above the previously encountered 10% limit and the saturation then occurs above 20% efficiency allowing the setup to reach the 25% max efficiency limit for the Gaussian pulse. As the XPW generated pulses in each crystal interfere constructively, 20% overall efficiency is reached with approximately 5% XPW efficiency in each crystal, to avoid any problems due to SPM.

Under these conditions, in our experiments, the XPW efficiency reached 15%, due to the not purely Gaussian spatial shape of the beam.

In both experimental cases, the input intensity on the crystal is moderated, and conservatively kept below the continuum threshold, i.e., about $10^{12}\,\mathrm{W\,cm^{-2}}$ [19]. Furthermore, in both cases, the cubic dependence on intensity is verified, which allows for the comparison between the experimental results and the theoretical model presented in the first section.

3.2 *Experimental results*

We first measured the chirp dependence of the spectrum after XPW generation using the Fourier limited 65 fs pulse. For each initial chirp value, the input energy is adjusted to keep the same energy density on the crystal. Consequently, the XPW efficiency is constant during the experiments. The spectrum of the XPW signal is registered for each initial chirp value and compared to the initial one (Fig. 3).

We have noticed that, whatever the initial chirp value we have applied during our experiments, the XPW spectrum exhibits a smooth Gaussian shape and is always broader than the initial one. The observed broadening does not change with the sign of the initial quadratic spectral phase, which is a strong indication of the limited influence of the nonlinear phase on the pulse broadening. But this broadening

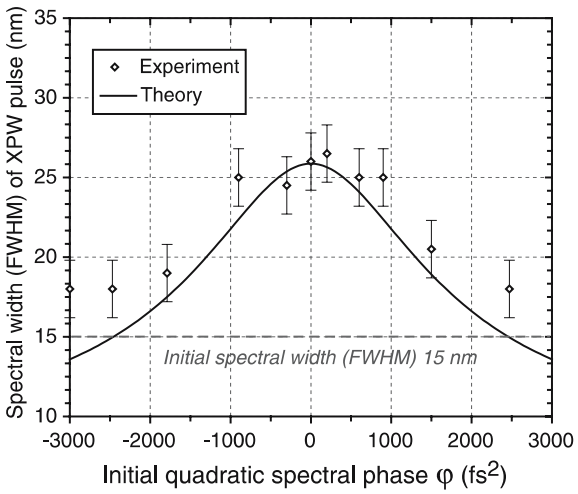


FIGURE 3 Measured spectral width (FWHM) of the XPW pulse (*squares*) as a function of initial quadratic spectral phase value compared with theoretical predictions (*full line*). The input pulse duration is 65 fs. The *dashed line* is the spectral width of the initial pulse

strongly varies with the phase value, as illustrated in Fig. 3. The best broadening is obtained for a null chirp value. In that case, the spectral FWHM of the generated pulse is about 26 nm compared to 15 nm before the nonlinear effect. The theoretical model developed in Sect. 2.2 has predicted the spectral width increase by a $\sqrt{3}$ factor when the chirp is negligible. Furthermore, thanks to this model, we have calculated the theoretical spectral width of the XPW signal corresponding to the experimental conditions. Equations (20) and (18) are used ($\Delta t_0 = 65$ fs, φ is variable). The Fourier transform limited duration of the XPW pulse ($\Delta t_{0\mathrm{XPW}}$) is thus determined, allowing for the calculation of the associated spectral width for a central wavelength $\lambda_0 = 820$ nm. Theoretical results, displayed in Fig. 3, show good agreement with experimental measurements.

To validate this behavior with shorter pulses and in a contrast filtering XPW generation setup (i.e., with higher efficiency of the overall XPW generation), additional experiments have been performed with the nonFourier limited 23 fs pulse. Those measurement have been performed at first in a vacuum to avoid SPM in air around the BaF_2 crystals.

Figure 4 shows the chirp dependence of the spectrum width after XPW generation with this shorter laser pulse. The observed XPW pulse spectra are Gaussian shaped. We observe the same symmetrical behavior of the spectral width around the null value of the pulse chirp. The width of the spectrum starts to reduce, compared to the useful input one, for a chirp value of around $\varphi = \pm 300\,\mathrm{fs}^2$. This result is consistent with the value for the 23 fs pulse from Fig. 2.

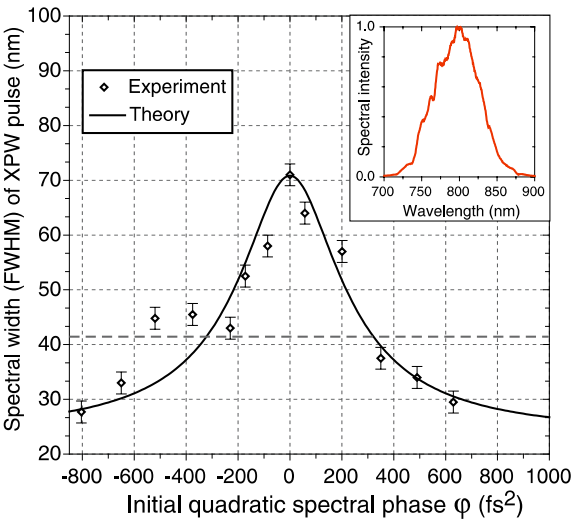


FIGURE 4 Measured spectral width (FWHM) of the XPW pulse (*squares*) as a function of initial quadratic spectral phase value compared with theoretical predictions (*full line*). The input pulse duration is 23 fs. The *dashed line* is the useful spectral width of the initial pulse. The XPW efficiency is 15% and the filter is in vacuum. (*Insert*) Experimental XPW spectrum measured when $\varphi = 0\,\mathrm{fs}^2$

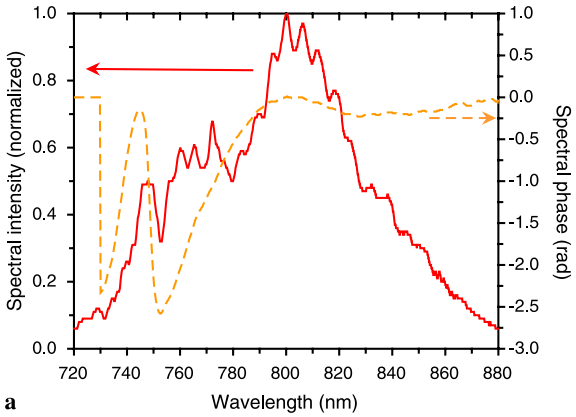
For the null chirp value, the spectral width is increased by a factor of $\sqrt{3}$ if we consider the useful spectral width giving a 23 fs Fourier limited pulse, i.e., 42 nm. The XPW generation process rejects amplitude modulations in the initial spectrum, as they are mainly responsible for the temporal side lobes. In that case, the broader spectrum measured (70 nm), corresponds to a 14 fs Fourier transform pulse. We then can no longer neglect the influence of the higher order spectral phase and amplitude modulation in the spectrum. But, as the 23 fs pulse behaves with the chirp according to the theory, we can dissociate the influence of chirp and the influence of higher order spectral phase on the spectral broadening.

As this high efficiency two-crystal configuration is the one of interest for contrast enhancement, it is interesting to discuss the experimental evidence of the eventual role of the nonlinear phase in the spectral broadening.

The first observation is that the spectrum in Fig. 4, typical of a XPW spectrum, exhibits a Gaussian shape including energy in the wings of the spectrum. According to [20], this profile should correspond to a nonlinear phase in a $\pi/2$–π radian range.

Actually, the self-phase modulation the XPW pulse undergoes is negligible because of the relatively low intensity of the signal itself. Consequently, to evaluate the nonlinear phase accumulated by the XPW signal, we have to consider the cross-phase modulation induced by the main pulse. By assuming a constant intensity of the fundamental pulse during propagation in the nonlinear medium, a nonlinear length of 4 mm, and a nonlinear refractive index of BaF_2 of $2.10 \times 10^{-20}\,\mathrm{m^2W^{-1}}$, the self-phase modulation quantity is 5 radians for the main pulse. This is an upper limit, as we do not consider fundamental depletion during the process. The resulting cross-phase modulation value on the XPW pulse is inferior to π radians. This value is not high enough to induce significant spectral broadening, as confirmed by the experimental behavior of the spectra described above.

Therefore, even in the high efficiency regime, the XPW process is the only one responsible for spectral broadening and associated pulse duration reduction.



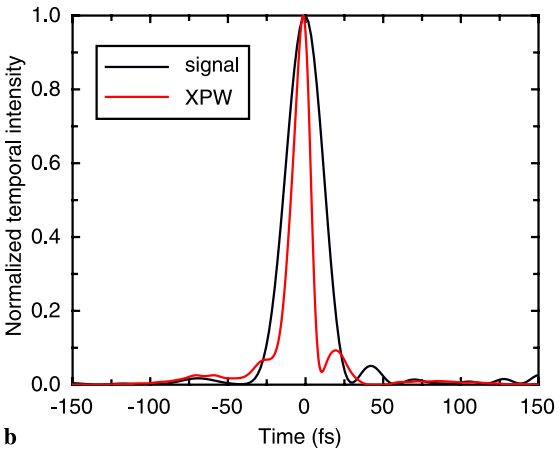


FIGURE 5 The input pulse duration is 23 fs and $\varphi = 0\,\mathrm{fs}^2$. The XPW efficiency is 15% and the filter is in air. (**a**) Measured XPW spectrum and SPIDER measurement of the XPW spectral phase. (**b**) SPIDER measurements of the temporal intensity of the initial pulse and the XPW pulse

As a consequence, it is clear that pulse shortening is compatible with temporal contrast improvement, as already demonstrated in [5]. Moreover, we can now explain that in early experiments [4], where a 10^{10} contrast pulse was measured, the spectral width had not increased because of the residual second order spectral phase introduced by optical elements (lenses, thick Glan polarizer) before the nonlinear crystal (about $1000\,\mathrm{fs}^2$, just below the critical phase value for 50 fs pulse, according to Fig. 2).

Finally, to evaluate the XPW temporal shape, we have performed SPIDER measurements of the XPW pulse for a null chirp value. The XPW pulse is compressed using a two-LaK8 prisms compressor to compensate for the dispersion introduced by the collimating lens and the output analyzer. Unfortunately, we have not been able to maintain the XPW under vacuum during this measurement.

Figure 5a shows the spectrum and spectral phase of the XPW generated pulse. Instead of the previously observed Gaussian pulse we have measured a strong modulation on the amplitude and the phase of the spectrum due to SPM in air around the focus spot of the 1500 mm lens. This long focal length, necessary for efficient XPW generation, provides a long enough Rayleigh range to promote a visible SPM contribution. A picosecond pedestal visible on the SPIDER could degrade the overall contrast enhancement due to XPW generation. We may conclude that SPM in air is the only process responsible for the modulated spectral shape shown in Fig. 5a, since during the previous XPW experiments performed under vacuum the measured spectrum had a Gaussian shape. Then, Fig. 5b shows the $\sqrt{3}$ reduction of the 23 fs pulse down to 14 fs. Moreover, the measured duration of 14 fs is coherent with the observed spectral behavior since spectral components due to SPM only contribute, in our case, to the temporal lobes generation. The registered pulse duration reduction demonstrates the validity of (16) and the interesting potential of the XPW process to shorten pulses even in the presence of a residual high order spectral phase.

4 Conclusion

In this paper, we have validated the calculation of duration, spectral phase and spectral width of a XPW generated pulse. The evolution of this pulse duration and spectral width depends strongly on the input pulse characteristics (duration and residual chirp). The optimal experimental conditions require a perfectly compressed input pulse. In that case, for a Gaussian spectrum, a precise control of the chirp allows for a pulse duration reduction by a factor of $\sqrt{3}$ and the associated spectral broadening during the process. Measurements with 65 fs and 23 fs pulse durations have experimentally validated our calculation allowing us to predict XPW pulse behavior. Moreover, 14 fs pulses have been generated by the XPW process. Further investigations of the effects of higher order spectral phases are underway.

Pulse duration reduction is an advantage of a XPW nonlinear filter for contrast enhancement. The filter transmission in terms of peak power is significantly increased, and allows for generation of shorter pulses with very high contrast or for compensation of spectral gain narrowing in the preamplifier.

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