# Efficient cross polarized wave generation for compact, energy-scalable, ultrashort laser sources

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**Abstract:** The generation of high contrast and ultrashort laser pulses via a compact and energy-scalable cross polarized wave filter is presented. The setup incorporates a waveguide spatial filter into a single crystal XPW configuration, enabling high energy and high intensity transmission, efficient contrast enhancement and pulse shortening at the multi-mJ level. Excellent XPW conversion of up to 33% (global efficiency: 20%, intensity transmission: 40%) led to an output energy of 650 µJ for an input of 3.3 mJ. Additionally, efficient conversion under specific input phase conditions, allowed pulse shortening from 25 fs to 9.6 fs, indicating the prospective application of this setup as a high energy, ultrabroad laser source.

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#### **1. Introduction**

Cross polarized wave generation (XPW) is a well established technique for contrast enhancement of ultrashort pulses in high energy laser systems. XPW is based on a degenerate four-wave mixing process governed by the anisotropy of the real part of a crystal's third-order nonlinearity tensor  $[\chi^{(3)}]$ , in which a new wave, polarized in the orthogonal direction, can be efficiently generated [1]. Implementation of XPW is quite straightforward: a linearly polarized laser pulse is focused into a crystal positioned in between two crossed polarizers. Efficient conversion with XPW only occurs at high intensities. Weaker, unconverted pre and post pulses are thus rejected by the second polarizer, thereby improving the temporal contrast of the pulse. Despite its simplicity, XPW has several drawbacks especially in terms of simultaneously achieving high conversion efficiencies and output energies. Limitations in seeding the nonlinear filter with high energies arise from the upper intensity limit of white light generation (<10<sup>13</sup> W/cm<sup>2</sup>) in the crystal (BaF<sub>2</sub>), while high conversion efficiencies require excellent beam quality. For a simple, single crystal setup, multi-mJ input energies could be used either by utilizing long focal lengths (>10 m) or working out of focus. Both solutions have tradeoffs: the first resulting to a bulky, extended setup while the second may lead to hotspots in the incident beam profile which is unfavorable for XPW, thus limiting the internal conversion efficiency to the order of 10-15% [2]. Dual crystal setups are capable of reaching efficiencies as high as 30% [2] yet the separation between the two crystals also depends on the focusing. Hence for multi-mJ pulses, the crystal distances lengthen to tens of meters, resulting in complicated and cumbersome configurations.

The most promising approach for simultaneously having high energy and highly efficient XPW resides in improving the efficiency of the single crystal setup. Smooth, flat-top like beam profiles are preferable for high conversion efficiencies, theoretically reaching up to 37% with Z-cut crystallographic orientation crystals [2] (45% with holographic-cut [3]), and in addition, higher energy transmission in the crystal is possible due to reduced self-focusing [3]. The conversion efficiency of a single crystal setup was improved to 28% for sub-mJ pulses with a sophisticated, nonlinear technique for beam shaping combined with pinhole filtering [3,4]. Energy scalability is possible but not simple due to the complexity of the technique incorporating nonlinear beam shaping. Implementation of a simpler setup at the multi-mJ level, relying solely on pinhole filtering, proved to be insufficient for efficient XPW generation. With a pinhole alone, the excellent efficiency primarily due to beam shaping was unobtainable and decreased drastically to less than 10% [5].

Due to these limitations, until now, XPW filters have only been suitable for the front-end of high-power laser chains at the sub-mJ level [6]. Contrast enhancement via XPW in these multi-terawatt lasers has proven to be crucial in particle acceleration [7,8]. Efficient XPW conversion at high input energies allows contrast enhancement at the final stage of the multi-mJ laser. This possibility is attractive since it eliminates the need of further amplification and

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provides a direct, high contrast laser source for intermediate energy experiments such as high harmonics generation.

Another interesting feature of XPW is that it permits spectral broadening of the pulse by at least a factor of  $\sqrt{3}$  [9]. Moreover, further broadening occurs at high intensities and high conversion efficiencies due to the interplay between cross phase and self phase modulation (SPM) of the fundamental and XPW pulses [3]. In line with obtaining high conversion efficiencies, XPW can be utilized as a tool not only for contrast enhancement but for spectral broadening as well.

In this paper, we demonstrate efficient XPW generation (33%) with high energy input pulses up to 3.3 mJ. The suggested configuration is based on a single BaF<sub>2</sub> crystal XPW stage and the use of a short length, hollow core waveguide as a spatial filter. The setup is established to be energy scalable, highly efficient, pulse shortening and carrier-envelope phase (CEP) stable. Up to 50% intensity transmission is also attained in the setup as a result of the efficient conversion and pulse shortening. The setup is simple, compact and flexible as it allows contrast enhancement for a seed beam of a laser chain or the final stage of a multi-mJ laser and a new device for spectral broadening.

#### 2. Experimental setup

In order to validate the concept, an initial study of the hollow core waveguide combined with XPW was first performed with a 1-kHz CEP stabilized laser system (Femtopower Compact Pro CE-Phase, Femtolasers GmbH) delivering 25 fs pulses at 1.5 mJ. An acousto-optic programmable dispersive filter (Dazzler, Fastlite) within the system allows the precise spectral phase control of the laser pulses. As a succeeding step, a demonstration of the scalability of this experiment was carried out using a similar laser with an additional multipass amplifier, boosting the available input energy to a maximum of 3.3 mJ and 30-fs pulse duration [10].



Fig. 1. Experimental setup of hollow core waveguide combined with XPW. Improvement of the beams incident on the  $BaF_2$  crystal is shown in the inset beam profiles of the non-filtered input and spatially filtered beam. The XPW output beam is also included.

The experimental setup is presented in Fig. 1. Linearly polarized input pulses are coupled by a 1.5-m focusing mirror into a 20-cm long, 250-µm diameter, fused silica hollow core waveguide with an efficiency of 85%. Beam pointing feedback is employed to stabilize the coupling into the waveguide. The filtered beam is then directly sent through a single 2.5 mm thick BaF<sub>2</sub> crystal ([011] crystallographic orientation) at a variable distance (25-41 cm) from the waveguide end. Both the waveguide and the nonlinear crystal are placed in the same vacuum chamber (sealed with two 500-µm fused silica windows) to avoid nonlinear effects in air. For the output analyzer, we either use a Glan or a thin film polarizer (Femtolasers GmbH). In the first case, the extinction ratio is better than  $10^{-3}$  and is degraded to about  $10^{-2}$ in the second case, facilitating however the compression of the XPW pulses with a set of chirped mirrors (Femtolasers GmbH).

The key design feature of this setup is the novel filtering concept of the input beam. Instead of simple spatial filtration as for a pinhole, filtering is based on a coupling process into the hollow core waveguide. Alignment of the waveguide predominantly supports the excitation of the fundamental mode while other modes are strongly attenuated due to higher losses. This effect removes any hotspots and non-uniformities of the input beam, producing a

smooth profile ideal for XPW generation even while working out of focus. The out of focus laser beam profile incident on the crystal is significantly improved with the waveguide as shown in Fig. 1. The filtering capacity of such a spatial waveguide filter is independent of the input beam profile and more tolerant to pointing instabilities of the input source. Pointing instabilities are transformed into coupling efficiency variations whereas for hard aperture filtering, they cause distortions of the beam profile by diffraction. At high input energies, it is difficult to improve the beam profile simply with a hard aperture because the filtering process occurs abruptly at the pinhole and requires complicated nonlinear beam shaping in the polarizer [4]. Both are required to obtain a super Gaussian profile thus achieving the best results [3]. On the other hand, filtering in the waveguide is gradual and based on modematching which, as a consequence, imposes the output mode. High energy pulses are spatially filtered over its length and not only at a single point. Additionally, the waveguide acts as a very convenient tool for the adjustment of the intensity level on the XPW crystal, permitting easy scalability of the input energy. In fact, the divergence of the filtered beam (~4 mrad) [11] allows the crystal to be positioned a few tens of centimeters after the waveguide, resulting to a very compact setup even for multi-mJ level inputs.



Fig. 2. (a) XPW internal conversion efficiency and output energy with respect to the input energy. (b) XPW spectral evolution with respect to the second-order phase at an input energy of 1.5 mJ. Internal conversion efficiencies are indicated on the left while spectral bandwidth (FWHM) values are on the right.

#### 3. Results and discussion

Figure 2(a) shows the evolution of the XPW energy and efficiency as a function of the input energy up to 1.5 mJ. The maximum output energy is 315  $\mu$ J corresponding to a 19.3% global throughput and a high internal XPW efficiency of 32%, accounting for losses from the windows, uncoated crystal surfaces and waveguide. Saturation of the efficiency to around 30% is observed for energies greater than 1.3 mJ. The energy stability of the input and XPW pulses has been measured to be 0.74% and 1.27% rms respectively.

The sensitivity of the XPW process versus the second-order spectral phase of the input pulses is shown in Fig. 2(b). As expected, the best spectral cleaning is attained close to the best conversion efficiency, as seen in Fig. 2(b). The rectangular-like spectrum of the input laser beam is smoothed into Gaussian-like and broad spectra for high efficiencies. Internal efficiencies above 20% are achieved for the variation of the second order phase within a range of  $\pm$  300 fs<sup>2</sup> around the optimum value. Spectral broadening via XPW up to about 100 nm is observed with the addition of positive chirp ( $\pm$  200 fs<sup>2</sup>). This is at the cost of some acceptable energy loss (27% efficiency) and a more rectangular-like spectral shape. Nevertheless, the result offers an interesting perspective towards the compression of short laser pulses directly from XPW generation. The conversion efficiency tolerance to third order phase variations is also measured to be within  $\pm$  2000 fs<sup>3</sup> around the optimum, signifying the consistency of the XPW conversion to variations in spectral phase [12]. We observe that in general, reasonable conversion efficiencies (>20%) are obtained effortlessly for a wide range of spectral phase values because with the waveguide filter, we can approach the theoretical efficiency of a single crystal XPW setup. This is a supplementary advantage since the setup can be

implemented together with laser systems without precise control or perfect optimization of the spectral phase of the laser pulse, which is crucial for broadband, few-cycle pulses.



Fig. 3. (a) Temporal characterization of the ultrabroad XPW pulse via FROG. Temporal profile and temporal phase with retrieved trace as inset (error = 0.10%) and (b) retrieved spectrum, measured spectrum and spectral phase. (c) Slow drift of the stabilized CEP of the XPW pulse at an input of 1.5 mJ.

To exploit the full capacity of our spectral broadening setup, we also experimentally studied the optimum combination of the spectral phase and the incident intensity on the crystal. In general, by simultaneously increasing the second order phase and intensity, an almost constant XPW efficiency (~30%) is achieved. The high intensity on the crystal induces SPM on the fundamental laser spectrum and therefore broadens the XPW spectrum as well. Optimization of the XPW spectrum could be achieved via second and third order phase scanning with the Dazzler, resulting in very broad spectra (>140 nm) having conversion efficiencies around 30%. The XPW pulses are compressed with chirped mirrors and measured via a single-shot frequency resolved optical gating (FROG) device (Fig. 3a-b). Optimum conditions yield pulse shortening from 25 fs to 9.6 fs (Fourier transform limit: 8.4 fs), broadening to 148 nm and an energy of about 300 µJ. It is probable that inhomogeneous spectral broadening along the beam profile may arise from SPM. The pulse duration at the center of the XPW beam may be shorter than the pulse duration at its periphery. The FROG measurement however, averages the pulse duration over the whole beam [13] which in the worst case, overestimates the shortest pulse duration along the beam profile. Likewise, the modulated spectrum is a characteristic sign of SPM. This slightly degraded spectral quality might induce some temporal defects on a picosecond time-scale but is an acceptable trade-off for the straightforward generation of ultrashort pulses. Contrast enhancement can be estimated as given in [2] to be from  $10^{-8}$  to 3.11 x  $10^{-10}$  due to the extinction ratio of the thin film polarizer. Pulse compression to the sub-10 fs regime solely via XPW has never been demonstrated before. With this capability, the setup may serve as a broadband seed source for the succeeding amplifiers of a laser chain or a new method for shortening a multi-mJ laser source. Lastly, with the shortening of the pulse, the setup reaches an impressive global intensity transmission of 50%.

Preservation of the CEP in a single crystal XPW setup has recently been confirmed [14]. The CEP stability of this new setup is examined using a small part of the compressed pulse directed to an f-2f interferometer and analyzed with a commercial software (Menlo Systems APS800). The error signal is fed back to the stretcher of the laser system to stabilize the overall slow phase drift of the laser amplifier and the XPW stage. As shown in Fig. 3c, the measured CEP deviation after XPW is ~0.33 rad rms over 120 s, not too far off from the typical rms CEP stability value of 0.2 rad for the laser alone (integration time: 1 ms, cycle loop time: 100 ms). Degradation of the stability is mainly due to air turbulence over the rather long, uncovered propagation path (~2 m) after the amplifier. We strongly believe that straightforward technical improvements on both issues would result in the enhancement of the XPW CEP stability level to that of the amplifier's.

To demonstrate the reliable energy-scaling ability of the setup, we performed an experiment with an input energy of 3.3 mJ coupled into the same waveguide. The position of the  $BaF_2$  crystal is adjusted to about 41 cm away from the waveguide end and optimized to

reach the highest conversion efficiency, not the shortest pulse duration. At this level, we achieve an output XPW energy of 650  $\mu$ J and record internal efficiency of 33%. Similar spectral behavior is observed, compared to the previous experiment at 1.5 mJ. Pulse shortening from 30 fs down to 15.5 fs is confirmed after compression, together with a good spectral quality (Fig. 4a-b). An intensity transmission of 40% is attained at this energy level. Sub-10 fs pulse shortening is possible as well by moving the crystal closer to the waveguide thereby inducing SPM and carefully optimizing the spectral phase.



Fig. 4. (a) Temporal characterization of the high energy XPW pulse via FROG. Temporal profile and temporal phase with retrieved trace as inset (error = 0.14%) and (b) retrieved spectrum, measured spectrum and spectral phase. (c) Temporal contrast of the laser at 3.3 mJ and XPW pulse measured with a third-order cross-correlator.

Temporal contrast is measured with a homemade third-order high-dynamical crosscorrelator [15]. The specific device has a 10-order of magnitude dynamic range when seeded with above 500  $\mu$ J. The cross-correlation is shown in Fig. 4c. The amplified spontaneous emission level of the laser is measured to be  $10^{-7}$ . Consequently, XPW improves this by 3 orders of magnitude down to  $10^{-10}$ , as determined by the extinction ratio of the Glan polarizer. The contrast could further be improved up to  $10^{-12}$  with the use of high quality polarizers (extinction ratio  $10^{-5}$ ). As seen in Fig. 4c, two pre-pulses are found in the XPW pulse: one inherent in the laser at -2 ps and effectively suppressed down to  $10^{-7}$  and one observed only in the XPW pulse at -7 ps, which may be an artifact since it disappeared in other measurements.

#### 4. Conclusion

In conclusion, we present the highest ever reported efficiency for single crystal XPW generation at the multi-mJ level. High energy and intensity transmission is realized accordingly from efficient XPW conversion and pulse shortening. The suggested design is based on a novel spatial filtering configuration resulting in a compact and energy scalable setup. Furthermore, the effective spectral broadening due to the efficient XPW process, allows the compression of the input pulse to sub-10 fs. We believe that implementation of the device as an add-on to existing multi-mJ laser systems will directly result in high energy, contrast ratio enhanced, pulse shortened and CEP stable ultrashort laser sources. The maximum energy limitation of the setup is only imposed by the damage threshold of the waveguide. Moreover, in the point of view of industrial applications or for robust and reliable laser chains, the setup is very interesting because of its simplicity and compactness. Owing to its scalability, such a source could either be directly used for multi-mJ level high intensity experiments or as a broadband seed source for further amplification in laser chains.

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