

# Highly efficient nonlinear filter for femtosecond pulse contrast enhancement and pulse shortening

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We propose a highly efficient scheme for temporal filters devoted to femtosecond pulse contrast enhancement. The filter is based on cross-polarized wave generation with a spatially super-Gaussian-shaped beam. In a single nonlinear crystal scheme the energy conversion to the cross-polarized pulse can reach 28%. We demonstrate that the process enables a significant spectral broadening. For an efficiency of 23% the pulse shortening is estimated to 2.2, leading to an intensity transmission of the nonlinear filter of 50%. © 2008 Optical Society of America

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The development of femtosecond petawatt-class laser systems relies on pulse energy increase and pulse duration reduction. At the same time, an excellent temporal quality is expected. In particular, critical care has to be devoted to temporal contrast improvement, with the implementation of suitable devices in the laser chain, directly at the output [1] or in a double-chirped-pulse amplification (CPA) configuration [2]. In both cases, such devices, besides contrast enhancement, are required to limit energy losses and at least to preserve the input spectral bandwidth. In this Letter we present a nonlinear solid-state temporal filter adequate to seed high-gain amplifiers for petawatt-class lasers in a double-CPA structure. A high energy transmission up to 28% is obtained with a simple device. Furthermore, a pulse duration reduction by a factor of 2.2 is demonstrated, leading to the production of 11.5 fs pulses. The nonlinear filter is based on a cross-polarized wave (XPW) generation process, a frequency degenerated third-order nonlinear effect occurring in  $\chi^{(3)}$  anisotropic crystals, such as barium fluorite ( $\text{BaF}_2$ ) crystals [3,4]. The ability of the process to improve the temporal contrast of ultraintense femtosecond pulses by several orders of magnitude has been demonstrated [5–7]. We investigate XPW generation in a single  $\text{BaF}_2$  crystal pumped with a smooth super-Gaussian spatial beam. We demonstrate a maximum efficiency of 28%. The significant pulse duration reduction is shown to be a consequence of this high conversion level. Experimental observations are in good agreement with theoretical predictions. We notice that the overall energy transmission of the filter is comparable to the one obtained with other nonlinear filtering techniques, such as elliptical polarization rotation in air [8]. However, this method has been shown to limit temporal contrast enhancement and to produce modulated spectra owing

to the strong self-phase modulation driving the process [9,10]. These drawbacks are overcome by the herein discussed method.

Experiments are performed with a millijoule-level, 25 fs, 10 Hz repetition rate Ti:sapphire laser. The laser design is based on a ten-passes amplifier pumped with 7.4 mJ (Surelite, Continuum). As the amplifier is saturated and pumped with a spatially flat-top beam the amplified beam exhibits a similar shape at the output of the amplifier. The compressed pulses seed the XPW device composed of a pair of crossed polarizers, focusing and collimating lenses with a 2 m focal length, and a  $\text{BaF}_2$  crystal (Fig. 1). The crystal is 2 mm thick with a holographic crystallographic orientation whose suitability for XPW process has recently been demonstrated [11]. The crystal is settled under vacuum to avoid additional nonlinear effects in air. The measured extinction ratio of the polarizers is  $10^{-4}$ . XPW generation has mainly been studied with spatial Gaussian shapes. Here, we aim at investigating the process behavior with a fundamental spatial intensity distribution appropriate for an improved efficiency (ideally a rectangular beam shape) [12]. Consequently, the  $\text{BaF}_2$  crystal is positioned in a plane after the focus (25 cm) where the amplifier output profile is imaged. The fundamental beam exhibits a modulated nearly super-Gaussian spatial profile. However, to optimize the nonlinear process and to avoid damages on the crystal, a smooth spatial shape is required. To remove high-frequency modulations, a filtering hole (500  $\mu\text{m}$  diameter) is inserted in the focus plane, before the

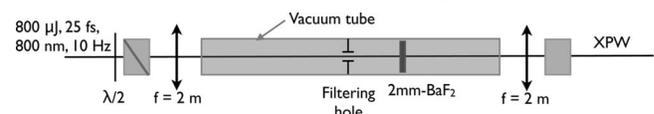


Fig. 1. Experimental setup.

BaF<sub>2</sub> crystal. This hole introduces 14% energy losses. The resulting spatial profile is smooth with an intensity distribution close to super-Gaussian (Fig. 2). The beam diameter on the BaF<sub>2</sub> crystal is estimated to 1.4 mm, allowing a maximum input energy of 800  $\mu$ J before the start of continuum generation in the nonlinear medium.

XPW energy conversion efficiency is measured as a function of input energy. Results are represented in Fig. 3. Efficiency curves are plotted as a function of the input energy and a parameter  $S$  convenient for calculations:  $S=(2\pi/\lambda)n_2I_0L$ . Experimentally,  $S$  is proportional to the intensity of the fundamental pulse and can be determined from experimental parameters. A record energy transmission, taking into account losses introduced by the hole and the uncoated crystal, is reached with this single-crystal scheme: 28%. So far, XPW internal efficiency in a single-crystal scheme has been limited to 15% in energy for the usual Gaussian spatial shapes, below the theoretical limit [12]. This limitation is due to intrinsic self-focusing in the nonlinear medium increasing self-phase modulation and then phase mismatch between the two waves (fundamental and XPW). This restriction is usually conveniently overcome by a double-crystal scheme, making possible an efficiency conversion above 20% [11–13]. In the case investigated here, a beam with a nearly rectangular spatial shape undergoes less self-focusing and consequently enables a high energy transmission with a single crystal. Experimental results are compared with a theoretical model developed with the solution of fundamental and XPW fields in holographic-cut crystals and presented in [11]. A calculation is run with the experimentally recorded fundamental spatial shape. The measured efficiency is in good agreement with theoretical dependence, until  $S\sim 3$ . After this value the experimental trend exhibits a saturation around 25%–28%—not predicted by the model. This early saturation is due to the nonpure rectangular spatial shape still weakly sensitive to self-focusing, a phenomenon that is not taken into account in the model. This self-focusing effect may be at the origin of fundamental pulse reshaping during propagation in the crystal and may induce phase-mismatching limiting the overall efficiency.

In our previous work, we demonstrated an attractive feature of the XPW process: pulse duration reduction and associated spectral broadening. In [14], a simple model estimates that at low conversion efficiency, when saturation effects are negligible, the XPW spectrum is  $\sqrt{3}$  broader than the input one.

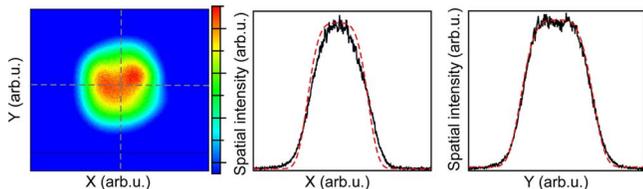


Fig. 2. (Color online) Experimental spatial profile of the input beam in the crystal plane after spatial filtering. Dashed curves, super-Gaussian fit ( $n=2$ ).

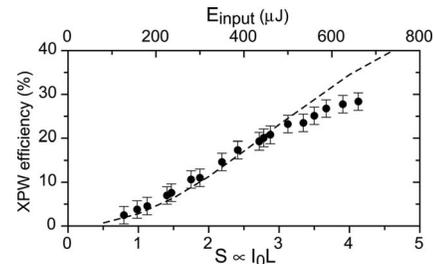


Fig. 3. XPW efficiency measured as a function of input energy, corrected for losses introduced by the filtering hole and uncoated faces of the crystal (filled circles). [ $S=(2\pi/\lambda)n_2I_0L$  with  $I_0$ , input intensity;  $L$ , crystal length;  $n_2$ , crystal nonlinear index]. Only the input energy is changed to modify  $I_0$ . Theoretical curve is calculated for a temporally Gaussian pulse and the experimental spatial profile (dashed curve).

Here, we show that high conversion efficiency enables one to exceed this  $\sqrt{3}$  factor. The experimental XPW spectral evolution as a function of process efficiency is displayed in Fig. 4(a). The XPW spectrum is uniformly broadened when the efficiency is increasing. Figure 4(b) represents the dependence of the output XPW spectral width (divided by the fundamental wave spectral width) on the process efficiency. The measured broadening factor is close to  $\sqrt{3}$  as far as the efficiency stays below  $\sim 15\%$ . Above this value, the XPW spectrum keeps broadening when the efficiency increases and can reach 2.5 times the initial spectral width. The developed theoretical model includes the spectral dependence (amplitude and phase) of both pulses during the nonlinear process. A calculation is performed to simulate experimental conditions by taking into account the real spectral intensity distribution and measured residual second- and third-order spectral phase of the input pulse. A super-Gaussian ( $n=2$ ) spatial distribution is considered. The estimated XPW spectral behavior appears in Fig. 4(b) (solid curve) and reproduces the experimental observations. This agreement indicates that the extra spectral broadening (above  $\sqrt{3}$ ) is due to four-wave mixing processes occurring during the interaction additionally to pure XPW process and included in the model: an interplay between cross-phase modulation and self-phase modulation of the XPW and fundamental waves. These effects and their spectro-temporal influence are not negligible when

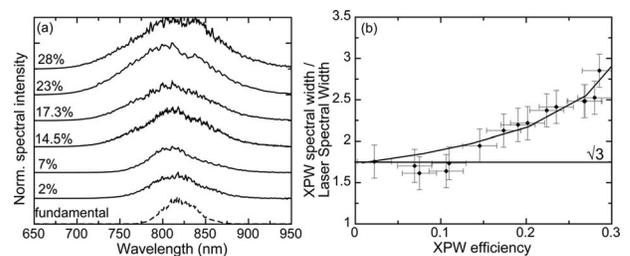


Fig. 4. (a) XPW spectral evolution and (b) XPW spectral width normalized by the input spectral width as a function of the XPW filter transmission (in energy, loss corrected). Theoretical spectral broadening calculated with experimental temporal parameters and a super-Gaussian ( $n=2$ ) spatial profile for the fundamental beam (solid curve).

the intensity of the XPW pulse increases (i.e., when the conversion efficiency is high).

It is noteworthy to underline that the broadened XPW spectrum presents an excellent quality: a smooth Gaussian shape, as shown in Fig. 5(a) (recorded for a XPW generation energy efficiency of 23%). This characteristic is necessary to preserve a good temporal quality and is a strong indication that the short XPW pulse can be fully compressed. To validate this last point, a spectral phase interferometry for direct electric-field reconstruction (SPIDER) measurement has been performed on the XPW beam without additional compression. A residual spectral phase ( $\phi^{(2)} = 1100 \text{ fs}^2$ ,  $\phi^{(3)} = 1900 \text{ fs}^3$ ) corresponding to residual phase of the laser and dispersion introduced by elements following the  $\text{BaF}_2$  crystal (window, lens, polarizer) is computationally removed. The result appears in Fig. 5(b): The XPW pulse can be compressed, for instance with a standard prism compressor, to a duration of 11.5 fs. The pulse duration is consequently reduced by a factor of 2.2 during the XPW process, which is consistent with the measured spectral broadening. This leads to an overall peak power transmission above 50% for this single-crystal XPW device.

Concerning the contrast improvement, the repetition rate of the laser and the energy level of the XPW signal are too low to perform high-dynamical third-order correlations. However, according to the knowledge that the contrast enhancement is limited only by the polarization discrimination, we are confident that the temporal contrast is improved by 4 orders of magnitude [12].

To conclude, we have demonstrated a high-efficiency XPW temporal filter based on a single-

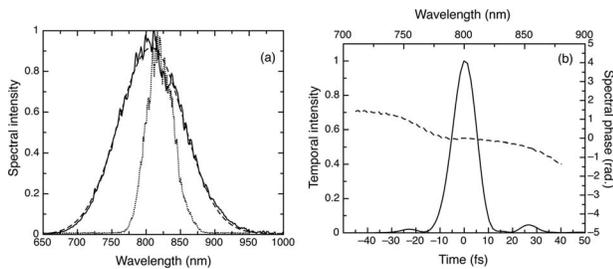


Fig. 5. (a) Typical XPW spectrum (solid curve) for 23% energy efficiency and fundamental spectrum (dotted curve). A Gaussian fit of the XPW spectrum is also represented (dashed curve). (b) SPIDER measurement: XPW temporal intensity and residual spectral phase.  $\phi^{(2)}$  and  $\phi^{(3)}$  corresponding to material before the apparatus (lens, polarizer, etc.) are computationally compensated. The pulse duration is estimated to 11.5 fs.

crystal device pumped with spatially super-Gaussian beams. The simplicity and high transmission of the setup combined with the significant pulse shortening would enable one to filter multimillijoule pulses. Indeed, in that case, high-peak power transmission is mandatory. Furthermore, a convenient way to filter energetic pulses is to place a single thick crystal in a collimated nonfocused beam to get the required intensity. The XPW device, combined with various beam-shaping techniques applied on the pump laser or on the infrared beam [15–17], could then be used to improve the contrast of multimillijoule pulses.

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