Broadband noncollinear optical parametric amplifier using a single crystal

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A noncollinear optical parametric amplifier in which blue pump pulse generation as well as parametric amplification takes place in the same nonlinear crystal is presented. Broadband spectra tunable throughout over 100 nm in the visible were generated in a simple setup. The green output pulses were compressed to 65 fs, and efficiency (IR-to-visible) of 1.8% was achieved. © 2005 Optical Society of America

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Sources of short, tunable laser pulses are essential to probe ultrafast processes in physics and chemistry. It was recognized in the early 1990s that a noncollinear geometry in parametric amplification matches the group velocity of the signal and idler pulses and thus allows nonlinear interaction between short pulses to take place in a relatively thick crystal.[1,2] Shortly thereafter the unique phase matching conditions for type I phase matching in β-barium borate (BBO) pumped by a frequency-doubled Ti:sapphire laser were discovered,[3,4] which led to the construction of what is now commonly referred to as a noncollinear optical parametric amplifier (NOPA). This source, commonly used in femtochemistry experiments, is now available commercially. The NOPA inherits the wide tunability of optical parametric amplifiers, at the same time allowing for broadband and efficient frequency conversion.

A typical NOPA setup driven by a Ti:sapphire regenerative amplifier consists of a second-harmonic generator that provides a strong (typically tens of microjoules) beam of ~100 fs pulses at ~400 nm, a supercontinuum generator, and an amplification stage based on a 1−2 mm thick BBO type I crystal. Pulses of several femtoseconds duration that are tunable throughout the visible and near IR[5] with microjoule energies have been reported. The output pulse energy can be further increased by appending a second amplification stage. Various improvements to the original NOPA scheme such as pump pulse tilting, supercontinuum precompression using prism dispersion lines and (or) broadband chirped mirrors as well as phase and amplitude modulators,[6–8] together with similar compression techniques applied to the amplifier output have led to the generation of pulses as short as 4 fs.[9] It was also recently demonstrated that broadband, high-efficiency parametric amplification of the supercontinuum can be realized in a strongly pumped long nonlinear crystal configured for second-harmonic generation.[10]

In this Letter we present a novel NOPA scheme, based on a marriage of second-harmonic generation and parametric amplification processes in the same nonlinear crystal (we call this a one-crystal NOPA).

The setup provides broadband tunable output with microjoule pulse energies.

The experimental setup of the one-crystal parametric amplifier is presented in Fig. 1. A train of 60 fs, 800 nm pulses from a chirped-pulse amplification system operating at 1 kHz was split on an uncoated glass plate. The transmitted beam (with up to 280 μJ energy per pulse) was focused with a 500 mm focal length lens into the BBO crystal (2 mm, $\theta = 29.2°$, p coated). The beam size on the crystal was measured to be 420 μm (FWHM), and the beam intensity was set just below the threshold for higher-order nonlinear interactions such as self-focusing and beam breakup. The crystal angle was first optimized for maximum second-harmonic generation, resulting in 35 μJ blue pulses. The beam reflected from the glass plate was attenuated and entered the supercontinuum generator where it was focused into a 3 mm thick rotating fused silica plate and the generated supercontinuum light was imaged onto the crystal. The pulse energy (within a 0.5−3 μJ range) and the position of the plate relative to the beam focus were chosen to provide stable, single-filament supercontinuum generation conditions. For sufficiently large IR pulse energies, apart from the second-harmonic beam, we also observed the cone of ampli-
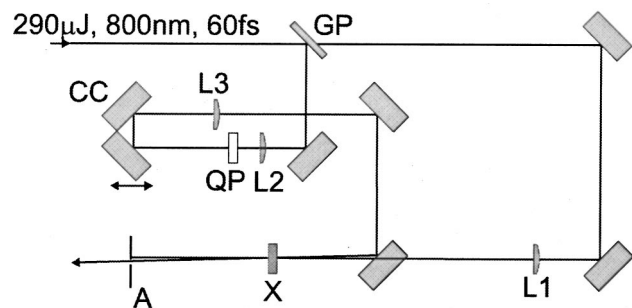

Fig. 1. Experimental setup of the one-crystal NOPA: GP, glass plate; L1, focusing lens, $f = 500$ mm; L2, focusing lens, $f = 120$ mm; L3, imaging lens, $f = 100$ mm; QP, 3 mm fused silica plate; CC, corner cube; X, 2 mm BBO crystal; A, aperture. White light generated in the glass plate travels below the figure plane after the first mirror.
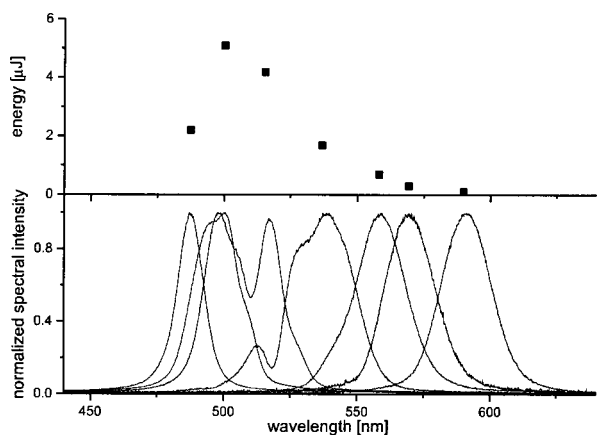
Fig. 2. Spectra of the amplified supercontinuum from the one-crystal parametric amplifier. Also plotted is the output pulse energy for different central frequencies.

fied spontaneous downconversion, emerging at 4.9° (half-angle, measured in air) from the BBO crystal.[10] The supercontinuum beam was aligned at an angle providing overlap with the cone in a way similar to that applied in the standard NOPA.[4] Amplified pulses were characterized using a powermeter (Scientech, Vector H-410) and a spectrometer (Ocean Optics, USB2000). While measuring the amplified pulse energies, we carefully subtracted the contribution from the IR pulses remaining in the supercontinuum beam.

The recorded spectra and pulse energies are presented in Fig. 2. We tuned the amplifier from 485 to 590 nm by changing the delay between the white-light seed and pump pulses and slightly adjusting the crystal angle. Throughout the whole tuning range the spectrum had a bandwidth exceeding 20 nm FWHM except at 485 nm, where the bandwidth was 11 nm. The maximum amplified pulse energy of 5.1 μJ was measured in the green around 505 nm. This value corresponds to 1.8% efficiency of IR-to-visible conversion. Measured peak-to-peak pulse energy stability was better than 10%, and the amplified beam maintained good spatial quality within the entire tuning range. With the spectrum centered at 525 nm we compressed the green pulses to 65 fs with a pair of fused silica Brewster prisms separated by 60 cm (pulse duration retrieved from intensity autocorrelation assuming a sech pulse shape).

The principle of operation of the one-crystal NOPA can be understood by taking into account two facts. First, one should realize that when a few-millimeters-thick nonlinear crystal such as BBO, phase matched for second-harmonic generation, is pumped with femtosecond laser pulses of the duration and energy typically found in regenerative amplifiers (several tens of femtoseconds and several hundreds of microjoules, respectively), a significant part of the second harmonic is generated in the first few hundred micrometers of the crystal. As a result, the blue pulse quickly acquires enough energy to pump the parametric process effectively in the remaining length of the nonlinear crystal. We have experimentally verified this by replacing the 2 mm long

crystal in our setup with a 0.5 mm crystal; the energy of the generated second harmonic dropped by less than 10% after such a swap.

The other important observation is that the phase matching angle for second-harmonic generation, $\theta_{SH,800\ nm} = 29.2°$, is close to the optimal pump angle for NOPA (i.e., the angle providing the broadest phase matching bandwidth for the signal beam). In a second-harmonic-pumped NOPA the crystals and beams angles are chosen to provide the same values of the projection of the signal group velocity and idler group velocity onto the signal beam propagation direction. Fulfilling this condition ensures broad amplification bandwidth, limited only by higher-order effects (mainly group velocity dispersion), around a certain central wavelength. As can be seen in Fig. 3, tuning the fundamental wavelength favors the amplification in a single crystal for various wavelengths: the second-harmonic phase matching curve crosses the optimum NOPA pumping angle curve for different signal wavelengths depending on the fundamental pump wavelength. In our case the bandwidth is expected to be broadest slightly above 500 nm. A similar observation can be made referring to Fig. 4, where we plot the angle between the signal and pump beams for perfect phase matching versus the signal wavelength. The amplification bandwidth in a given direction is broad in the region where such a tuning curve becomes horizontal. In Fig. 4 this happens for a signal wavelength around 505 nm and indeed the broadest spectrum and the highest amplified pulse energy were measured there. From Fig. 4 one can also see that for a fixed pump-signal angle there is a finite tuning range (defined by a phase matching curve and broadened by the angular spread of the pump beam and the crystal length)—amplification of longer wavelengths is possible if the pump-signal angle is changed.

We have demonstrated efficient operation of a one-crystal noncollinear parametric amplifier. In the simple setup, spectra that were tunable from 485 to 590 nm were generated with a bandwidth exceeding 20 nm and energies of a few microjoules. Compression of the green pulses to 65 fs with a prism
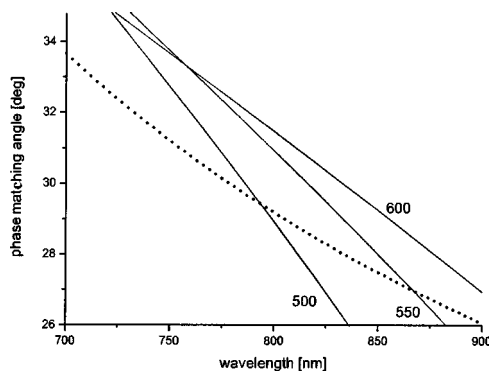

Fig. 3. Phase matching angle in type I second-harmonic generation in BBO versus fundamental wavelength (dots). The solid lines represent the optimal crystal orientations for pumping at the second harmonic of the horizontal axis wavelength for broadband noncollinear parametric amplification around 500, 550, and 600 nm.
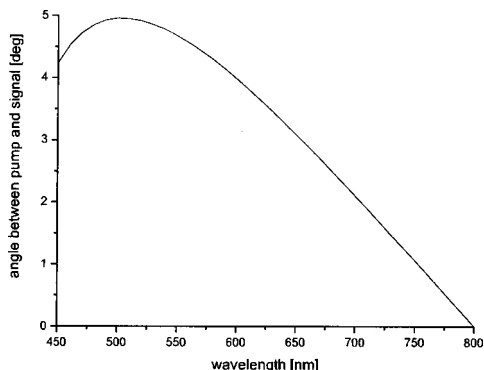
Fig. 4. Angle between the pump and signal beams for perfectly phase matched amplification in BBO versus signal wavelength. The crystal is oriented for collinear generation of the second harmonic from the fundamental beam centered at 800 nm.

pair was presented. Our device may be further developed to benefit from white-light supercontinuum pulse shaping and postamplification pulse compression. It is also possible to pump a second stage of the amplifier with the remaining blue light, thus increasing the output energy.

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