# Generation and amplification of ultrashort shaped pulses in the visible by a two-stage noncollinear optical parametric process

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We report the generation and amplification of ultrashort shaped pulses in the visible by a two-stage noncollinear optical parametric amplification process. Phase and amplitude profiles of the shaped pulses are conserved in our amplification scheme. The energy losses normally associated with the production of complex shaped pulses are eliminated. 

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Ultrashort laser pulse shaping is of growing interest in a wide variety of optical applications, including quantum and optimal control, high-speed communications, and material characterization.[1,2] Pulse shaping is usually done as a final step, although it usually introduces significant energy losses. Amplification of shaped pulses has been demonstrated based on a Ti:sapphire chirped-pulse regenerative amplification process.[3,4] but this limits the spectral range of the amplified shaped pulses to ~800 nm. If ultrashort shaped pulses are to be utilized in a wider range of applications and systems, methods need to be developed to generate shaped optical pulses at widely tunable wavelengths.

The recently developed noncollinear optical parametric amplification (NOPA) has provided an ideal tool for generating ultrashort pulses throughout the visible regime.[5–7] These light sources have been used in feedback-controlled pulse-shaping experiments to automate pulse compression.[8]

Preliminary studies of the amplification of shaped pulses by the NOPA process (by shaping of the white-light continuum before amplification) gave visible optical pulses with simple phase details.[9] In this Letter we show that one can significantly enhance this method by modifying a two-stage NOPA process,[10] whereby the first stage NOPA generates a stable pulse, which is then shaped before being amplified again by a second stage NOPA with a stretched pump pulse. This scheme produces sufficient intensity in the preamplified shaped pulse to allow its phase and amplitude profile to be conveniently measured. Direct comparison of the pulse shapes before and after the second-stage amplification process can then be made. This comparison allows us to determine whether the pulse shape is adequately conserved in the process. The ease of such comparison facilitates the generation of stable amplified shaped pulses in the visible with much more-complex phase and amplitude modulation.

A schematic of our experimental setup is shown in Fig. 1: 120-fs pulses centered at 800 nm (1-kHz repetition rate with a peak-to-peak fluctuation of 2%) from a regenerative amplifier system pump our two-stage shaped pulse generator and amplifier. This fundamental beam is split into three portions. One portion (120 $\mu$J) is used in the pulse shape characterization, as described below. Another portion (~10 $\mu$J) is used to generate a stable white-light continuum by focusing into a 2-mm-thick sapphire plate. The major portion (680 $\mu$J) is used in a second-harmonic generation process [type 1, 0.5-mm $\beta$-barium borate (BBO) crystal], generating pulses of 190 $\mu$J centered at 400 nm. These pulses are split almost evenly to provide the pump pulses for the first- and second-stage NOPA processes. The first-stage amplification process follows closely the prescription offered by Wilhelm *et al.*[5] The generated white-light continuum is focused into a 2-mm BBO crystal, whereas the pump pulses at 400 nm (~90 $\mu$J) are focused to a point 20 mm in front of the BBO crystal with a spherical mirror. The energy of the resultant pulses ranges from 13 $\mu$J at 630 nm to 9 $\mu$J at 515 nm.

These first-stage NOPA pulses are then steered into an acousto-optic pulse shaper (AOPS). The AOPS setup[2] includes a pair of 1200-groove/mm diffraction gratings and a pair of spherical mirrors ($f = 200$ mm) set up in a 4-$f$ configuration. The input pulse is shaped in the Fourier plane by a $TeO_2$ crystal (42 mm long, 10 mm thick). The specific geometry of our pulse-shaper setup provides a spectral window of

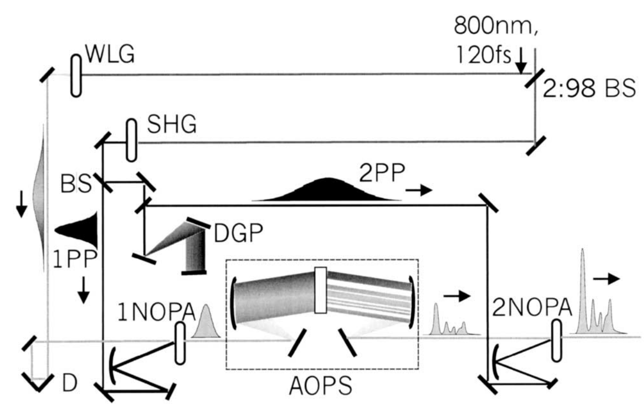


Fig. 1. Experimental setup for the amplification of shaped pulses by a two-stage NOPA: BSs, beam splitters; WLG, white-light generation (sapphire); SHG, second-harmonic generation (type 1, BBO); D, delay; DGP, diffraction grating pair; 1PP, 2PP, first- and second-stage pump pulses, respectively; 1NOPA, 2NOPA, first- and second-stage noncollinear parametric amplifiers (type 1, BBO), respectively.

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130 nm. The typical power throughput of the AOPS assembly is approximately 5–10%. The efficiency of the gratings and our intention to work in the linear regime of the acousto-optic effect account for this attenuation.

The spectra of pulses from the first-stage NOPA are measured, and an amplitude mask is produced to match the experimental spectrum with the desired shaped pulse spectrum. Unmodulated pulses are characterized to yield the amount of chirp as a result of material dispersion of the pulse shaper (typical values are group-velocity dispersion, $\sim 2000\ \mathrm{fs}^2$, and cubic dispersion, $\sim 22{,}000\ \mathrm{fs}^3$). Precompensation for this phase chirp is programmed into the AOPS, together with the desired phase profile and the amplitude mask mentioned above to generate the desired preamplified shaped optical pulse.

The preamplified shaped pulses are used to seed a second-stage NOPA system. Except for the duration of the pump pulses, the setup of the second stage NOPA is similar to that of the first. Shaped pulses are in general not transform limited and hence are longer than transform-limited pulses of similar spectral bandwidth. Amplifying these shaped pulses requires that the pump pulses be temporally longer. Hence a diffraction grating pair (600 grooves/mm) is used to stretch the pump pulses from ~150 to ~800 fs. The losses in the diffraction and steering process attenuate the pump pulses, originally at 100 $\mu$J, to 30 $\mu$J per pulse.

We characterize both the preamplified and the amplified shaped pulses with a variant of the spectrally and temporally resolved upconversion technique (STRUT).[11] A portion of the fundamental source provides the reference pulse (FWHM, 8 nm; centered at 800 nm). Spectra of sum-frequency generation (type 1, 80-$\mu$m BBO crystal) of the shaped pulse and of the reference pulse are obtained as a function of delay between the two pulses. The resultant time–wavelength correlation or STRUT trace gives the plot of group delay versus frequency component of the shaped pulse. By plotting the maxima of the sum-frequency spectra as a function of delay, we obtain the first derivative of the pulse phase with respect to angular frequency $\phi'(\omega)$.

Using the setup described, we are able to generate and amplify pulses with various complex shapes. Figure 2 shows a hyperbolic secant pulse with a hyperbolic tangent frequency sweep as an example. This type of pulse is of particular interest, as it provides an analytical solution to the optical Bloch equation in inverting the part of the population of two-level systems that falls within a resonance window.[12,13] The field envelope of this pulse can be described by

$$E(t) = \mathrm{sech}(\alpha t)^{1+\mu i}, \qquad (1)$$

where $\alpha = (0.57 \times 300\ \mathrm{fs})^{-1}$ determines the pulse duration and $\mu = 21$ parameterizes the frequency sweep. The theoretical pulse intensity $I(t)$ and the first derivative of the pulse phase with time $\dot{\phi}(t)$ are shown in Fig. 2(a). The theoretical STRUT trace is depicted in Fig. 2(b). We generate the shaped pulse centered at 625 nm. The spectrum of this pulse resembles a rectangular profile with a width of 50 nm [Fig. 3(b)]. The experimental STRUT traces of the preamplified shaped pulse and of the amplified shaped pulse are shown in Figs. 2(c) and 2(d), respectively. The measured pulse energy of the preamplified shaped pulse is 0.15 $\mu$J. On amplification, the pulse energy becomes 4.8 $\mu$J with a pulse-to-pulse energy fluctuation of ~5%.

Figure 3(a) compares the first derivative of the phase with respect to the angular frequency $\phi'(\omega)$ of preamplified and amplified shaped pulses (retrieved from the STRUT trace) with theory. From the

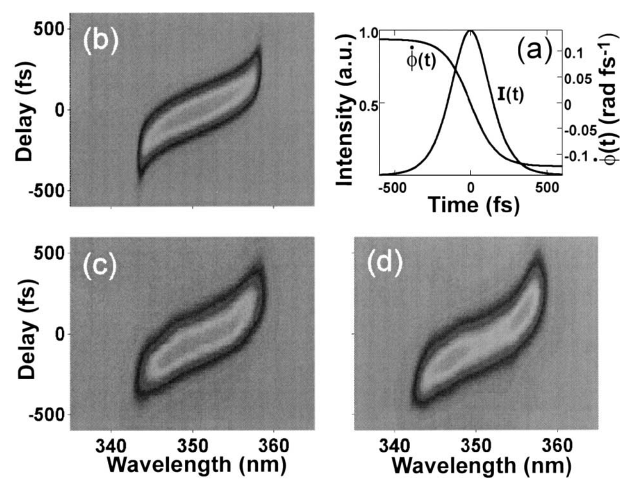


Fig. 2. (a) Theoretical plot of the intensity $I(t)$ and first time derivative of phase $\dot{\phi}(t)$ of a $\mathrm{sech}(\alpha t)^{1+\mu i}$ pulse in time, with $\alpha = (0.57 \times 300\ \mathrm{fs})^{-1}$ and $\mu = 21$. (b)–(d) Sum-frequency generation spectra versus delay (STRUT trace) of the shaped pulses (centered at 625 nm) with a reference pulse (centered at 800 nm). (b) Theoretical and (c), (d) experimental preamplified and amplified pulses, respectively.

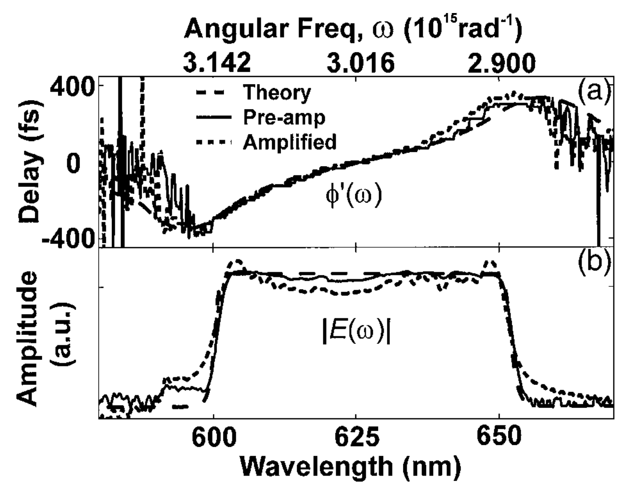


Fig. 3. (a) Comparison of the theoretical first derivative of phase $\phi'(\omega)$ and the experimental values recovered from the STRUT traces in Figs. 2(c) and 2(d) for preamplified and amplified $\mathrm{sech}(\alpha t)^{1+\mu i}$ pulses, respectively. (b) Comparison of the electric field amplitude $|E(\omega)|$ obtained theoretically and that of the experimental values, obtained from the spectrometer for the preamplified and amplified shaped pulses.

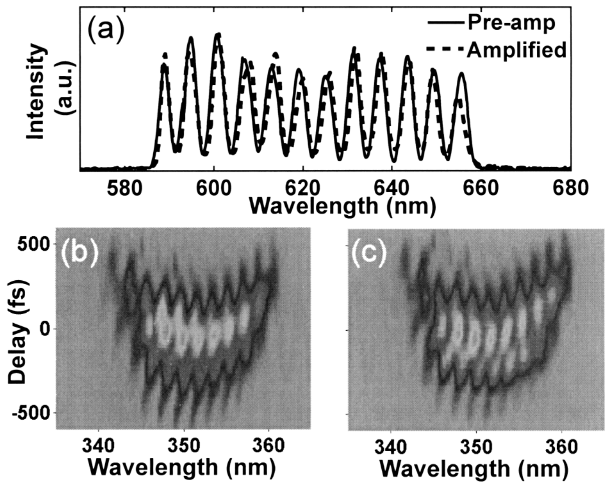


Fig. 4. (a) Comparison of the spectra of preamplified and the amplified shaped pulses with a sine-squared amplitude modulation across the 70-nm spectrum. Experimental STRUT traces of (b) preamplified and (c) amplified shaped pulses with a cubic chirp of 30 000 $fs^3$.

comparison we can see that, besides showing good agreement with theory, the complex phase profile of the shaped pulses is adequately preserved in the noncollinear optical parametric amplification process. Figure 3(b) compares the experimental electric field amplitude in the spectral domain of the preamplified and amplified pulses obtained by a spectrometer with the theoretical amplitude profile.

We also demonstrate (in Fig. 4) amplification of a shaped pulse with strong amplitude modulation in the spectral domain. We start with a pulse with a rectangular spectrum of width 70 nm centered at 625 nm with a cubic phase chirp of $30,000\ \mathrm{fs}^3$. We then impose an amplitude modulation function of $\sin^2(\beta\lambda)$ in the wavelength domain, with $\beta = 0.51\ \mathrm{nm}^{-1}$. The preamplified pulse is amplified from 0.1 to 4 $\mu$J by the second-stage NOPA process. From a comparison of the spectra of the preamplified and the amplified shaped pulses in Fig. 4(a) we observe that the amplitude modulation of the shaped pulse is well preserved in the NOPA process. Figures 4(b) and 4(c) compare the STRUT traces of the preamplified and amplified shaped pulses and show that the phase profile (cubic phase chirp of $30,000\ \mathrm{fs}^3$) is preserved adequately as well.

In the present study, for the second stage NOPA process, pump pulses with a duration of 800 fs suffice to amplify shaped pulses of duration ~400 fs. The upper limit imposed on the duration of the amplified shaped pulses increases with the temporal length of the pump pulses as long as the pump pulse's peak intensity exceeds the threshold needed to sustain the nonlinear optical process. Modern commercial Ti:sapphire regenerative amplification systems are capable of delivering as much as 3 mJ of ultrashort pulses at a 1-kHz repetition rate. With the experimental minimum value of pump pulse peak intensity needed to sustain the NOPA process taken from recent literature,[14] pump pulses as long as 10 ps can still be expected to sustain the NOPA process for our present setup, and several-picoseconds-long shaped pulses can be adequately amplified. It should also be noted that the quadratic phase chirp of the pump pulses that is due to the stretching process does not distort the phase details of the amplified pulse, as only the absolute value of the temporal pump pulse profile matters. The ~5-$\mu$J energy per pulse output with a 30-$\mu$J pump is comparable in efficiency with NOPA processes reported in the visible.[5–7] One can increase the final amplified shape pulse energy by increasing both the pump pulse energy and the efficiency of the pump pulse's stretching process.

In summary, we have demonstrated, for the first time to our knowledge, the amplification of nontrivially shaped pulses by a noncollinear optical parametric process with the conservation in its phase and amplitude profile. Shaped pulses with complex phase structure (e.g., the hyperbolic tangent frequency sweep) and strong amplitude modulation (e.g., sine-squared function) centered at 625 nm with a bandwidth of as much as 70 nm are amplified from 0.1 to ~5 $\mu$J. Generalization to shaped pulses centered at other wavelengths in the visible is straightforward and is restricted only by the tunability of the NOPA system.

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