# Shaped ultrafast laser pulses in the deep ultraviolet

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**Abstract:** We use an acousto-optic pulse shaper to directly control the phase and amplitude of femtosecond laser pulses in the deep ultraviolet ($\sim$ 260 nm). The efficiency of the pulse shaper is 21% and the output pulse energy is 2.8$\mu$J. We are currently using these pulses in molecular coherent control experiments.



**OCIS codes:** (320.5540) Pulse shaping; (140.7090) Ultrafast lasers.

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## 1. Introduction

There is substantial interest in using ultraviolet light for probing molecular chemistry on a femtosecond timescale. Optical pulse shaping (for example see [1, 2, 3]) has been used extensively in applications that require control over broadband laser pulses. By combining pulse shapers and feedback algorithms [4], a variety of novel control experiments have been performed [5]. Recently, pulse shaping has been extended into the ultraviolet using a micromirror SLM [6] or a fused-silica acousto-optic modulator (FS-AOM) [7, 8], both operating at the second harmonic of a Titanium:sapphire laser (near 400 nm). Shaped, ultrashort pulses in the ultraviolet have also been produced by transferring shaped pulses in the visible or near infrared to the ultraviolet via non-linear frequency mixing techniques (for example, see [9, 10]). In this paper, we extend direct pulse shaping of broadband femtosecond pulses into the deep ultraviolet ($\sim$ 260 nm). Shaping the ultraviolet (UV) after generation allows for high-energy shaped pulses (few microjoules), while maximizing fidelity. An advantage of working in the ultraviolet with an acousto-optic pulse shaper is that wavelength-dependent diffraction effects are much less important than in the infrared because the range of wavelengths required to support a given spectral bandwidth is proportional to the central wavelength.

## 2. Experimental set-up

We begin with amplified femtosecond pulses from a standard Titanium:sapphire laser system, producing pulses at 785 nm with 30 fs duration and 1 mJ of energy at a repetition rate of 1 kHz. Approximately 700 microjoules of the light is down-collimated into a 250 micron thick BBO crystal cut for second-harmonic generation of the fundamental, producing 175 microjoules of light at 393 nm. Since the group velocity inside the BBO crystal is wavelength dependent, the peak of the second harmonic pulse exits later in time that the fundamental. To compensate for this mismatch, both the fundamental and second harmonic pulses propagate through a 1 mm thick calcite crystal (cut at $\theta = 41^{\circ}$) oriented such that the group velocity mismatch is reversed, allowing the 393 nm pulse to catch up to the 785 nm pulse. Finally, light at the third harmonic (262 nm) is produced via sum-frequency-generation of the 785 nm and 393 nm light in a 100 micron thick BBO crystal. With our current non-linear crystals and focusing geometry, we produce 13 microjoules of light in the deep ultraviolet.

The UV light passes through a standard 4-f based pulse shaper with 3600 g/mm aluminum gratings and 750 mm focal length dielectric mirrors. For the shaping element, we use a 20 mm FS-AOM (Brimrose) that is computer-controlled via an arbitrary waveform generator (GaGe) synthesizing shaped, radio-frequency waves directly at 150 MHz. This arrangement allows for complete control over both the phase and amplitude of the laser pulses. Given the 1 GHz sample rate of the waveform generator and the 50 MHz bandwidth of the AOM (FWHM), we are able to specify the phase and amplitude of the wave at approximately 170 points across the AOM aperture. With our current gratings and mirrors, the optical bandwidth does not entirely fill the acoustic aperture, limiting us to approximately 150 independently addressable frequency components.

Since the diffraction efficiency of the FS-AOM is quite good as these wavelengths (we achieve 78% into first order with 2 W drive power), the output energy of the shaped 262 nm light is 2.8 $\mu$J. The shaper efficiency (21%) is currently limited by grating losses with the P-polarization required for the FS-AOM. Since the gratings are relatively independent of polarization at these wavelengths, rotating the polarization between the gratings and AOM would

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only increase the shaper efficiency by approximately 25%. The pulses are measured using the technique of self-diffraction (SD) FROG [11], where a 250 micron thick sapphire plate serves as the non-linear optical element.

The gratings in the pulse shaper currently limit the shaping to pulses with central wavelengths above 240nm. Replacing the gratings could extend the use of the pulse shaper to pulses with central wavelengths as low as 200 nm, at which point the absorption of the FS-AOM becomes a problem.

## 3. Results

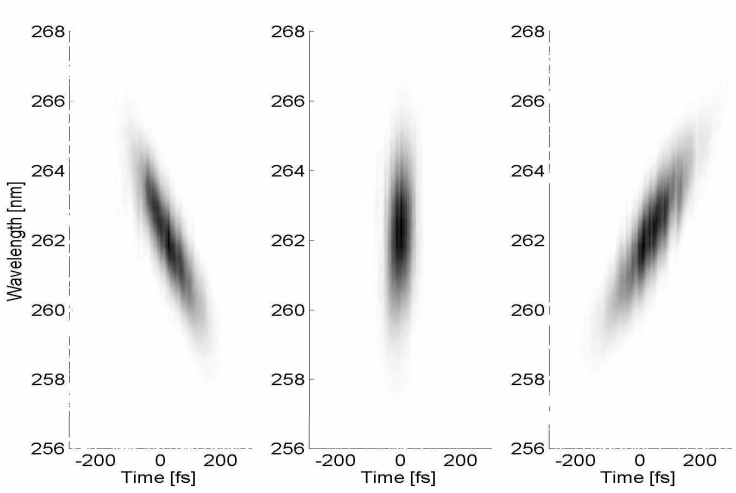


Fig. 1. SD-FROG plots of positively-chirped (left), unshaped (center), and negatively-chirped (right) ultraviolet pulses created by programming quadratic spectral phase onto the AOM.

Figure 1 shows SD-FROG plots of positively-chirped (left), unshaped (center), and negatively-chirped (right) pulses. The pulses are created by programming quadratic spectral phase profiles onto the AOM. The pulse durations (FWHM) are approximately 120 fs for the chirped pulses and 55 fs for the unshaped pulse. The bandwidth-limited duration of the pulse is approximately 50 fs. This pulse duration is currently limited by the frequency tripling process, as we choose non-linear crystals that provide higher pulse energies but sacrifice some bandwidth. The measured FROG traces agree well with predictions based on the phase programmed onto the acousto-optic modulator.

As an example of a more complicated pulse that the shaper can produce, Fig. 2 plots the SD-FROG of a laser pulse with a sinusoidally-modulated phase profile. This phase for this pulse corresponds to a sine wave with approximately 0.22 oscillations per THz of bandwidth and a depth of modulation of 2 ($-1$ to $+1$) radians. The characteristic pulse train (with an intra-pulse spacing of 220 fs) produced by sinusoidal phase modulation is evident from the FROG measurement.

We have already begun to use these shaped deep-UV pulses in molecular coherent control experiments. The energies are sufficient to drive multiphoton dissociative ionization in several molecules, and the pulses are therefore well suited for direct control over molecular fragmentation. We plan on using the pulses for closed-loop control over molecular photodissociation (for example, see [12, 13, 14]) and isomerization [15].

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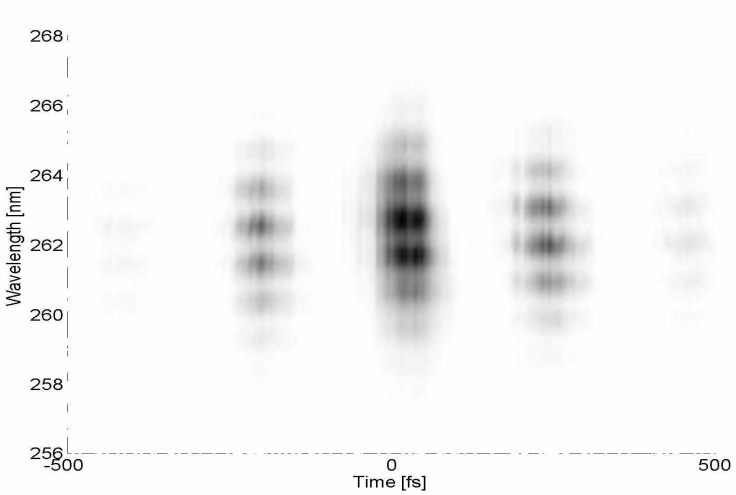


Fig. 2. SD-FROG plot of a pulse with sinusoidally-modulated phase.

## 4. Conclusion

We directly shape the phase and amplitude of femtosecond laser pulses in the deep ultraviolet ($\sim$ 260 nm) using a FS-AOM. The shaping technique maintains microjoule energies, allowing the pulses to be used in molecular coherent control experiments. We gratefully acknowledge support from the National Science Foundation under award number 0555214.

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