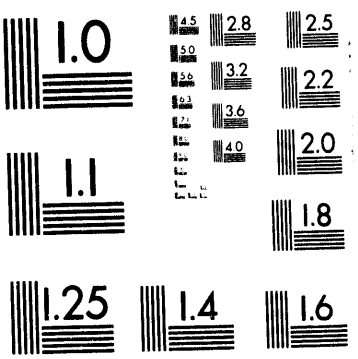
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Title: Ti:SAPPHIRE BASED ULTRAFAST PUMP-PROBE LASER SOURCE IN THE VIOLET AND ULTRAVIOLET

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Ti:sapphire based ultrafast pump-probe laser source in the violet and ultraviolet

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ABSTRACT

Frequency conversion of an ultrafast 810 nm Ti:sapphire laser has produced 15 mJ, 165-fs pulses at 405 nm with an energy conversion efficiency of 35%. Upon tripling the fundamental to 270 nm, an energy of 0.8 mJ is obtained. Unconverted 405-nm light is used to generate an ultrafast supercontinuum in water that extends from 200 nm to 600 nm. The system operates at a 5 Hz repetition rate, and is useful for applications in deep ultraviolet pump-probe ultrafast spectroscopy.

1. INTRODUCTION

Recent reports of frequency conversion of ultrashort, terawatt class Ti:sapphire lasers have demonstrated their usefulness in producing femtosecond sources of light in the ultraviolet (UV)[1,2] and near-vacuum ultraviolet (near-VUV)[3] wavelength range. The attraction of Ti:sapphire lasers for such a purpose is motivated by the development of Ti:sapphire as a robust solid state material as a femtosecond oscillator, and the property of Ti:sapphire as an efficient medium for joule level amplification. Coupled with the conversion efficiencies that nonlinear crystals can provide, the ·e of Ti:sapphire as a source for UV femtosecond lasers becomes attractive. In addition to using these sources to seed ·e rare gas halide excimer amplifiers in the ultraviolet[1,2], generation of wavelength tunable femtosecond laser ·es in the UV and near-VUV region[3] are practical for a number of applications including: subpicosecond UV k·· ·· spectroscopy[4,5], laser generated target plasmas[6], wide band gap semiconductors[7], and insulator material dynamics.[8]

For time resolved femtosecond pump-probe spectroscopy, often one relies on the absorption of a broadband probe continuum for the detection of a transient species spectral feature. Generally, the probe continuum is generated by an intense femtosecond source through a nonlinear process in a gas or condensed phase medium called supercontinuum generation.[9] This paper describes a femtosecond pump-probe system useful for detection of photochemical UV transients. The scheme for such a system relies on frequency tripling an ultrashort Ti:sapphire laser to generate pump pulses at 270 nm. The UV probe continuum is produced by focusing intense violet femtosecond pulses at 405 nm in water to generate a continuum that spans a wavelength range of 200 nm to 600 nm. It is believed that this is the largest spectral coverage achieved with an ultraviolet/visible continuum to date.

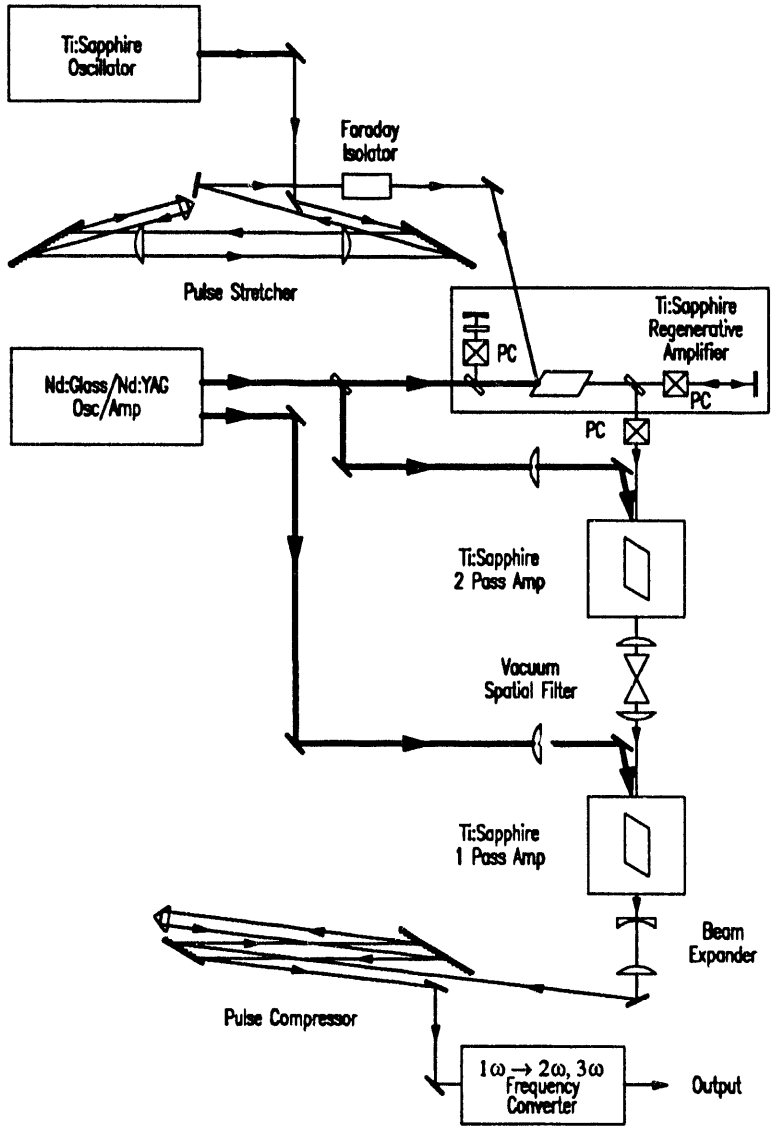

Figure 1. Optical layout for the Ti:sapphire laser system. The output wavelength before frequency conversion is 810 nm. The output energy and pulse width at 810 nm is 43 mJ and 125 fs, respectively. The system operates at a repetition frequency of 5 Hz.

2. Ti:SAPPHIRE LASER SYSTEM DESCRIPTION

The Ti:sapphire system layout, equipped with an oscillator and several amplifiers, is shown in Figure 1. The system is seeded with pulses from a commercial mode-locked Ti:sapphire laser operating at 810 nm and pumped by the 10 Watts, all-lines output of an Ar-ion laser. These 70-fs, 15-nJ pulses have a spectral bandwidth of 9.6 nm. Before amplification, these pulses are temporally stretched using a grating pair pulse-stretcher configured to yield positive group velocity dispersion.[10] Two antiparallel 2000-line/mm gold-coated holographic diffraction gratings are separated

by 1.7 m, and a unit magnification telescope using 60-cm-focal-length achromatic lenses is inserted between the gratings. After two passes through the stretcher, the measured pulse duration is 440 ps. Next, the pulses traverse a Faraday isolator. Losses in the beam up to this point (mostly in the stretcher) result in a 5-nJ/pulse energy to seed the amplifier chain.

The Ti:sapphire amplifier chain boosts the energy to 95 mJ and consists of a regenerative amplifier, a double-pass preamplifier and a final amplifier. All amplifiers are pumped using frequency-doubled Nd:glass/Nd:YAG oscillator/amplifier system.[11] The intensity of these 110-ns, 0.53-μm pump pulses are more than an order of magnitude smaller than those obtained from conventional Q-switched Nd:YAG lasers typically used to pump Ti:sapphire amplifiers, making the system more resistant to optical damage from the pump beam. The strategy used to increase the pump pulse duration is to utilize the low gain of Nd:glass in the oscillator to yield long-pulse-duration seed pulses at 1.062 μm, and then use Nd:YAG, with its superior thermal properties, for amplification up to joule-level energies. The laser system, described in detail in Reference 11, consists of a Q-switched glass oscillator, followed by three Nd:YAG amplifiers operating at a 5 Hz repetition rate. Frequency-doubling of the output with conversion efficiencies approaching 50% is achieved in two output arms using a 60-mm-long and a 100-mm-long KD*P crystals.

The first amplifier is a regenerative amplifier using a linear cavity with one flat end mirror and one 3-m focal length end mirror separated by 145 cm to form a stable TEM_{00} mode with a waist size of 1.63 mm located at the flat end mirror. The 7-mm-diameter by 20-mm-long Ti:sapphire crystal with 0.15% Ti doping yields a small signal gain of 4 when pumped with 65 mJ of 532-nm light focused to a diameter of 2.0 mm. The seed beam is input via a Brewster's reflection off of the Ti:sapphire crystal. A combination of two Pockel's cells, a quarter wave plate and a thin film polarizer is used for pulse selection and switch-out. A 5-ns, 3.6-kV pulse from a pulse generator switches the Pockel's cell. A total of 20 round trips in the cavity is required to boost the pulse energy from 5 nJ to 7 mJ.

The pulse is then amplified up to the 60-mJ level in a double pass Ti:sapphire preamplifier. The 7-mm-[illegible]meter by 20-mm-long, 0.15% doped, Ti:sapphire rod is pumped with 250 mJ of 532-nm light focused to a 3.0-mm-[illegible]meter spot. After the first pass the output energy from the Ti:sapphire beam is 28 mJ, and after the second pass, the double pass gain is 8.6 to yield pulse energies of 60 mJ. At the output of the preamplifier, the diameter of the Ti:sapphire beam is 2.9 mm. In order to improve the spatial quality of the output from the Ti:sapphire preamplifier, a vacuum spatial filter is placed between the preamplifier and the final amplifier. The vacuum spatial filter consists of a 125-μm-diameter diamond pinhole that has a 75% energy throughput efficiency including reflection losses from telescope lenses and vacuum port windows. The output energy after the vacuum spatial filter is 45 mJ. The telescope for the vacuum spatial filter is also used to expand the Ti:sapphire beam diameter to 4.2 mm before seeding the final amplifier.

The final amplifier consists of 12-mm-diameter by 20-mm-long, 0.15% doped, Ti:sapphire rod which is pumped with 400 mJ of green from the second output arm of the 532-nm pump laser. The single pass gain of the final amplifier is 2.6, yielding an output energy of 110 mJ for the Ti:sapphire beam. The beam diameters for the pump and the Ti:sapphire beam at this point are 4.4 mm and 4.2 mm, respectively. Before injecting the amplified Ti:sapphire pulses into the grating compressor, the beam diameter is expanded and collimated to a diameter 9.3 mm with a dual lens telescope in order to minimize any potential for optical damage to the compressor gratings. After the telescope, the Ti:sapphire pulse energy is 95 mJ.

After, the Ti:sapphire amplifier chain, the pulse width is compressed by a factor of 3200 with a parallel grating pair. The compressor consists of a pair of 2000 lines/mm gold-coated holographic gratings separated by 74 cm. Although compressed pulse widths of 110 fs are achievable, day-to-day operation of the system yields a typical compressed pulse width of 125 fs. The throughput energy efficiency of the grating pair compressor is 45%, and the output pulse energy of the compressed beam is 43 mJ. Figure 2 displays the single shot autocorrelation after compression, and Figure 3 displays the final amplified Ti:sapphire output spectrum. The temporal and spectral widths in Figures 2 and 3 lead to a time-bandwidth product of $\Delta\nu\Delta t = 0.443$. Upon measuring the output energy stability at a repetition frequency of 5 Hz, the pulse-to-pulse amplitude stability is within 12%.

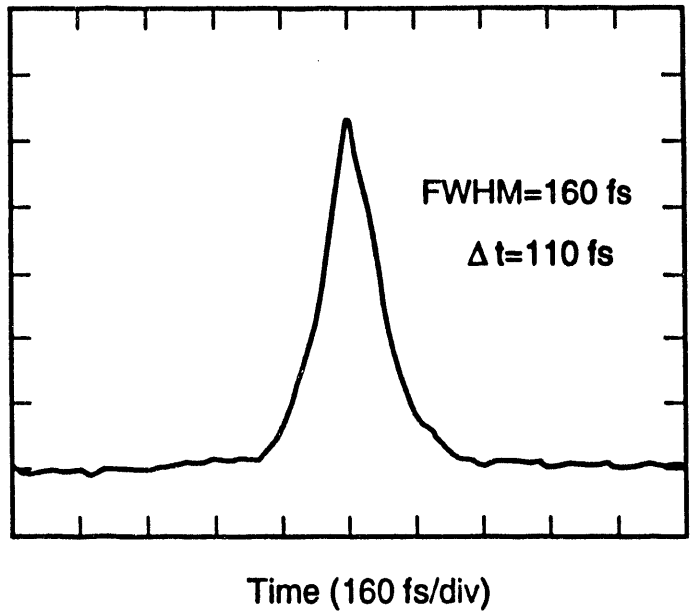

Figure 2. The single shot autocorrelation trace of the compressed Ti:sapphire pulses is shown. The FWHM is 160 fs, yielding a pulse width of $\Delta t = 110$ fs. Typical day to day operation of the laser yields pulse widths that are slightly longer, 125 fs to 160 fs.

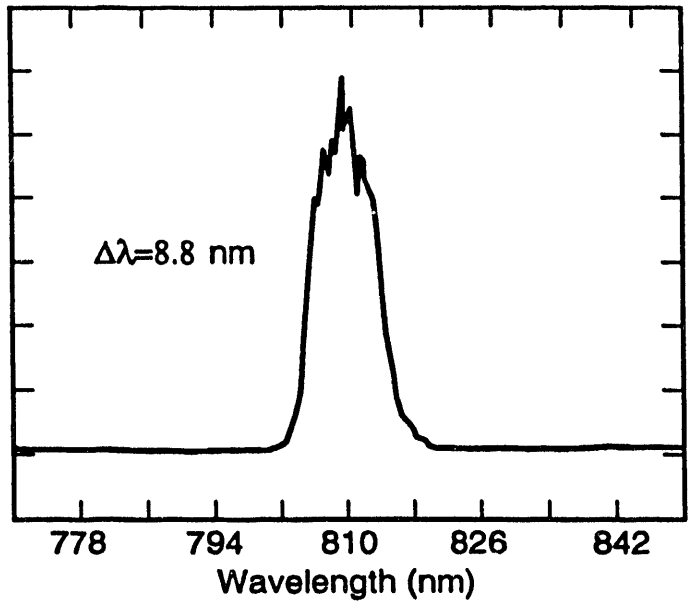

Figure 3. The ouptut spectrum of the amplified Ti:sapphire laser. The center wavelength is 810 nm, and the bandwidth is $\Delta\lambda = 8.8$ nm. Using the pulse width obtained from Figure 2, a time bandwidth product of $\Delta\nu\Delta t = 0.443$ is obtained.

3. FREQUENCY CONVERSION AND CONTINUUM GENERATION

Interest in the frequency conversion of such a system to the violet and ultraviolet has spawned experiments in using amplified ultrashort Ti:sapphire pulses for seeding krypton fluoride (KrF)excimer amplifiers at 248 nm after frequency tripling the fundamental wavelength of Ti:sapphire.[1,2] For the system described here, the calculated group velocity mismatch between ω and 2ω is 80 fs/mm, and the calculated group velocity mismatch between ω and 3ω is 390 fs/mm. A 1.5 mm thick KDP Type I crystal is used for frequency doubling to 405 nm, and a 0.5 mm thick KDP Type I crystal is used for frequency tripling to 270 nm. In order to avoid optical damage to the crystals, both crystals have large 35 mm diameter apertures, and both are Sol-Gel AR coated. Figure 4 shows the optical layout used for SHG Type-I and THG Type-I of the Ti:sapphire laser output using KDP. A delay line is employed to optimize the THG output energy. For SHG, an output of energy of 15 mJ at 405 nm is achieved. The SHG energy conversion efficiency is 35%. For THG, an output energy of 0.8 mJ at 270 nm with a conversion efficiency of 2% is obtained. With an input energy fluence of 63 mJ/cm^2, the output energy fluences for SHG and THG generation are 22 mJ/cm^2 and 1.2 mJ/cm^2, respectively. Figure 5 displays the output spectrum at 405 nm after SHG. The spectral bandwidth for the 405-nm pulse is $\Delta\lambda$=3.0 nm. The inset to Figure 5 shows the intensity crosscorrelation between 810 nm and 405 nm. Deconvolution of the crosscorrelation trace with a 160-fs 810-nm pulse reveals that the 405-nm pulse width is 165 fs. The time-bandwidth product for the 405-nm pulse is $\Delta\nu\Delta t$=0.903, which is twice the transform limit for Gaussian pulses. Figure 6 displays the output spectrum for THG generation. The spectral bandwidth of the 270-nm pulse is 1.5 nm. Although we are yet unable to measure the pulse width at 270 nm, it is expected that these pulses are sub-200 fs since the inputs, 810 nm and 405 nm, each have pulse widths under 165 fs.

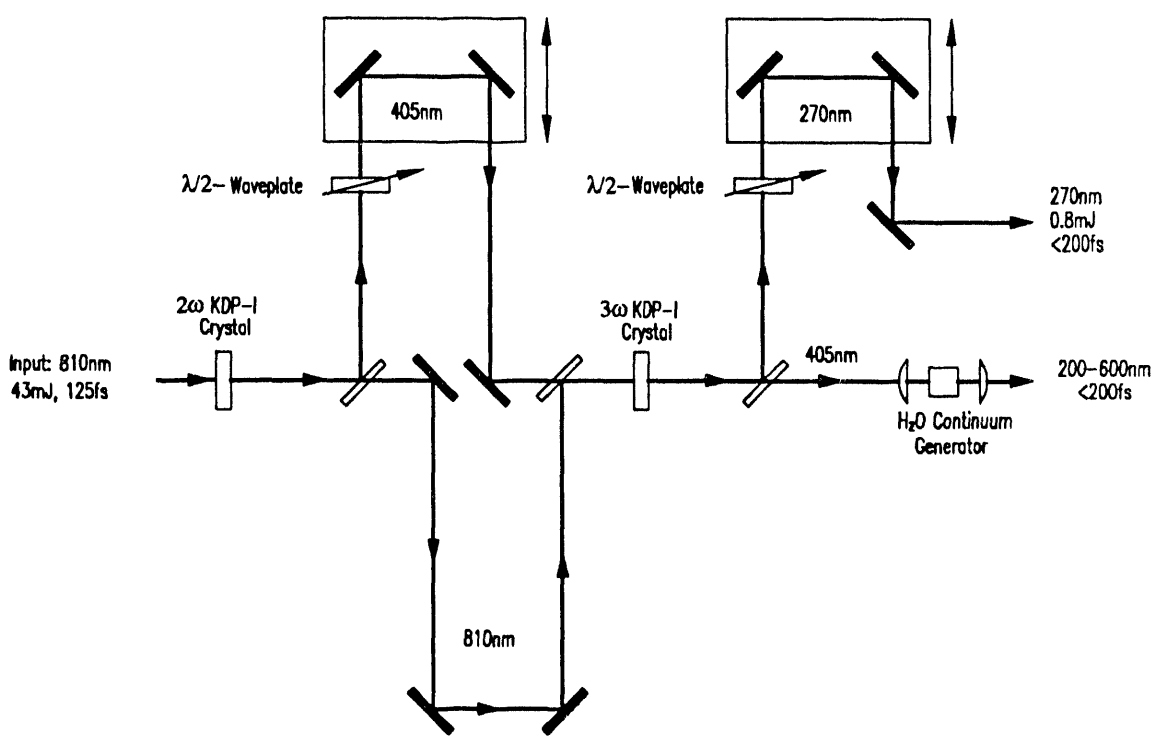

Figure 4. Optical layout for SHG, THG, and supercontinuum generation of the ultrafast Ti:sapphire laser beam.

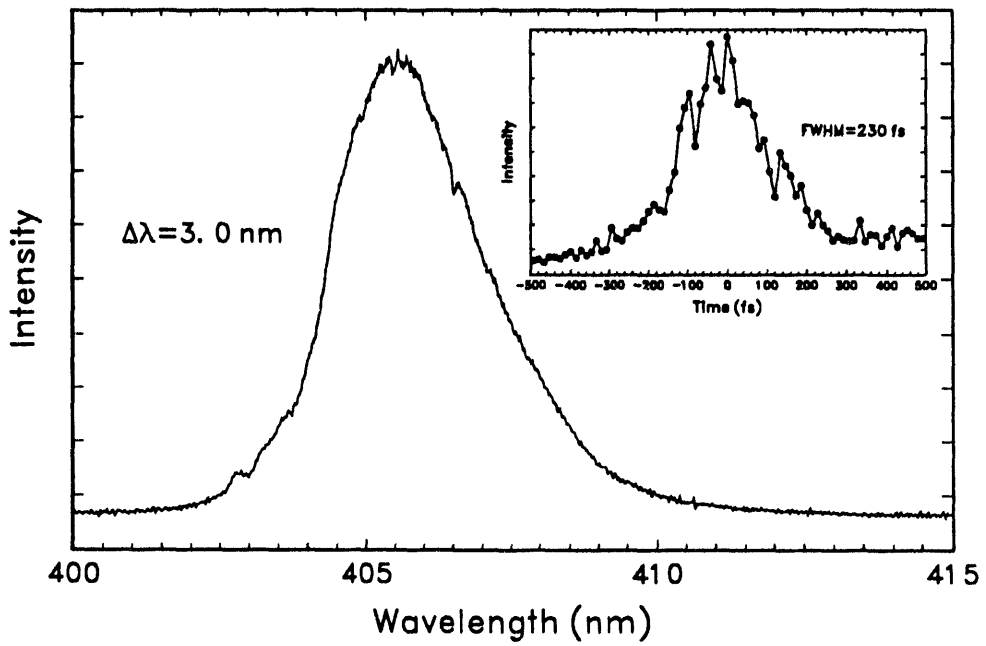

Figure 5. The SHG output spectrum at 405 nm. The bandwidth at 405 nm is $\Delta\lambda$ = 3.0 nm. The inset shows the intensity croscorrelation between the 810-nm and 405-nm pulses. The FWHM of the croscorrelation trace is 230 fs. After deconvolution, the pulse width at 405 nm is Δt_{405} = 165 fs. The 810 nm input pulse width is Δt_{810} = 160 fs.

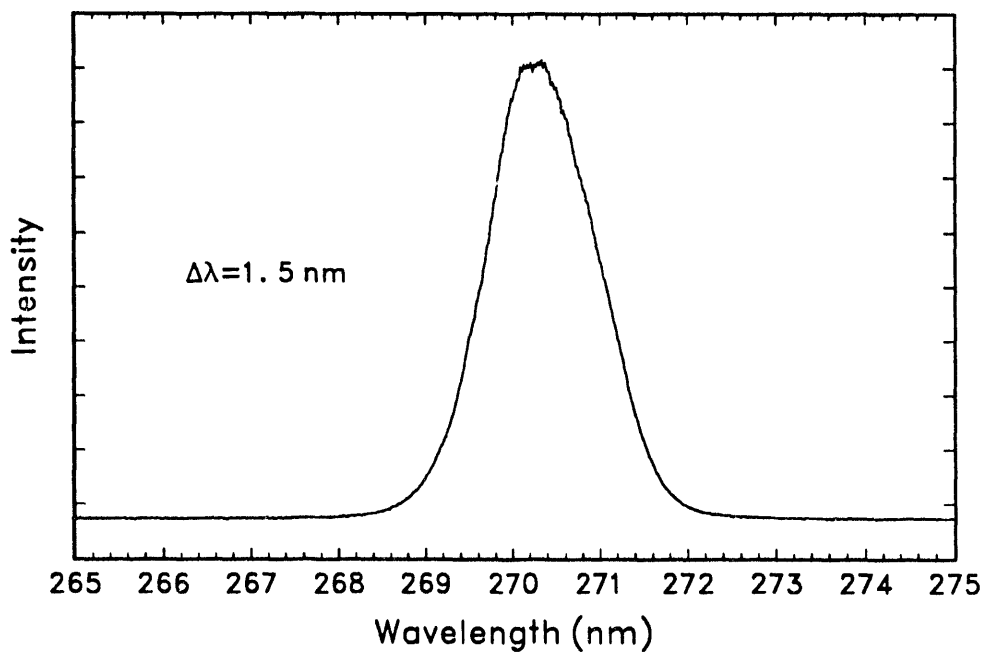

Figure 6. The THG output spectrum at 270 nm. The bandwidth at 270 nm is $\Delta\lambda$ = 1.5 nm. Given the bandwidth at 270 nm, and the input pulse widths at 810 nm and 405 nm, the pulse width is calculated to be <200 fs.

In addition to SHG and THG, Figure 4 also shows an additional delay line inserted into the system after frequency conversion of the Ti:sapphire beam. For sub-picosecond kinetic UV spectroscopy, the system is configured as a pump-probe apparatus. The 270-nm beam is utilized as an ultraviolet pump pulse, and after spectral filtering, the unconverted 405 nm light is used generate an supercontinuum in water (H_2O) for the probe. The time delay between the pump and the H_2O continuum probe is controlled using an optical delay line in the pump beam arm. Approximately 5 mJ of the 405-nm beam is used to focus into a quartz cuvette cell (path length = 5 mm) containing a static fill of H_2O. Figure 7 shows the spectral extent of the supercontinuum. Centered at 405 nm, the UV continuum extends from below 200 nm to beyond 600 nm. This is believed to be the shortest wavelength generated with an H_2O continuum to date. The extreme UV limit of the continuum is due to the loss of transmission in the quartz cell. The threshold energy for generating continua was measured to be 200 μJ. Figure 8 displays the H_2O continuum spectra between 270 nm and 280 nm obtained with five successive shots when pumping with 5 mJ at 405 nm. It shows that the shot-to-shot intensity fluctuation in the continua remains to within 20% of each other. The quality of the continuum can also be measured by the smooth structureless features that it displays. The smoothness of the continua throughout the 200 nm to 600 nm range is in contrast to the spectral modulation that may appear in ultraviolet supercontinua of rare gases[12,13]. Almost no micro-filamentation is observed in the H_2O continuum beam, and this is manifested in the good shot-shot amplitude stability. Finally, since this pump probe system operates at a repetition frequency of 5 Hz, [illegible]-to-pulse signal averaging should improve the minimal detectable absorption sensitivity to under a few percent.

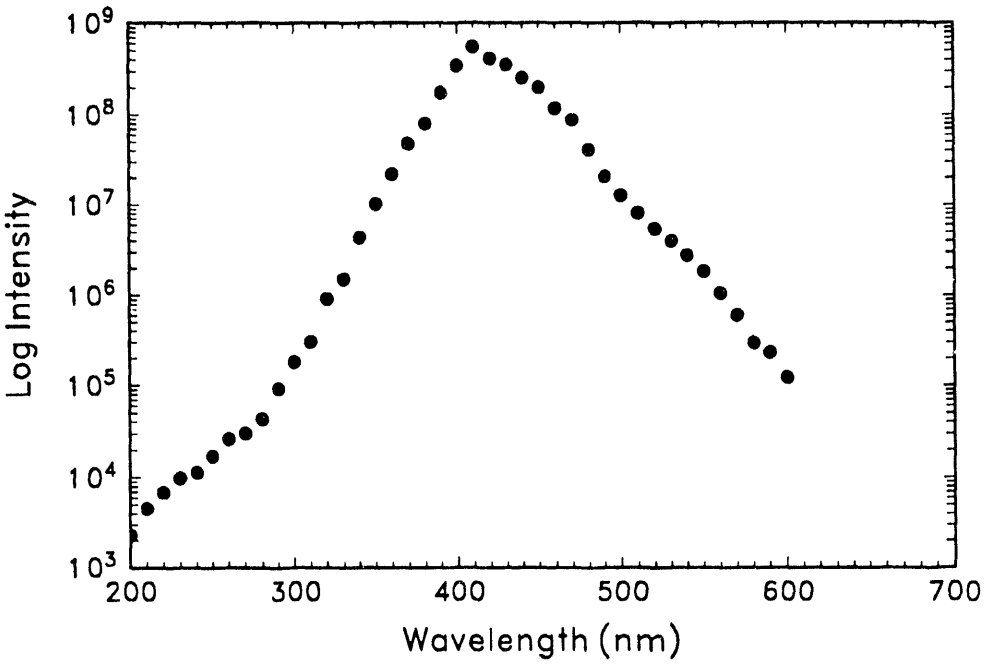

Figure 7. Plot of the water (H_2O) supercontinuum spectrum. The 405 nm input pulse energy and pulse width are ~5 mJ and 165 fs, respectively. The H_2O cell transmission is 80% at 220 nm.

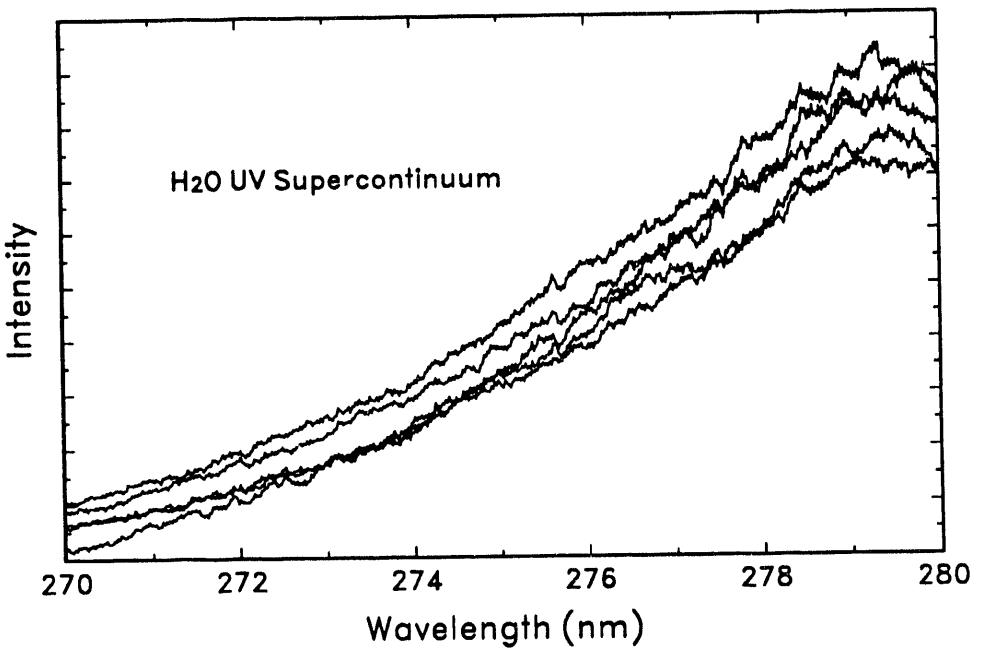

Figure 8. Supercontinuum spectra obtained with water in the 270 nm to 280 nm region. Five successive single shot spectra are displayed. The spectra illustrate good shot-to-shot amplitude stability with no spectral modulations.

4. CONCLUSIONS

In this paper the description and performance of a frequency doubled and tripled, 0.3 terawatt, 125-fs Ti:sapphire laser is discussed. With a 35% energy conversion efficiency, the output at 405 nm is 15 mJ with a pulse width of 165 fs. After tripling the Ti:sapphire wavelength to 270 nm, an output energy of 0.8 mJ is attained. The energy at 270 nm should be useful in serving as an ultrafast pump source for UV photolysis of molecules in a subpicosecond pump-probe apparatus. A tunable broadband probe source is demonstrated by generating a supercontinuum in water with unconverted 405-nm light after frequency tripling. The continuum extends from below 200 nm to beyond 600 nm, and appears to be smooth and free of any spectral modulation. The system should be useful in detecting deep UV photochemical transients in the subpicosecond time domain.

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