Ultra-short Milijoule-Level Amplification in Ti:Sapphire at 1 kHz and Third Harmonic Conversion Measurements

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ABSTRACT

We report on a Ti:sapphire amplifier system which produces 21 fs, 1 mJ pulses at 1 kHz. A nanojoule-level seed pulse makes 8 passes through a highly doped Ti:sapphire crystal for a total gain of ~10^6. The crystal is pumped with 12 mJ of 527 nm light which is converted into amplified pulse energy with an efficiency exceeding 10%. We have used this laser system to generate ultra-short UV light pulses through third harmonic generation. By focusing the 21 fs pulses in argon and other gases (including air), we obtain UV pulses with energy 1 μJ and wavelength 267 nm. The duration of the UV pulses was measured to be ~16 fs, representing the shortest pulses in the this wavelength range measured to date.

Key Words: Laser Amplifier, Titanium-Sapphire, Ultrashort Pulses, Third Harmonic Generation, Ultraviolet Light.

1. INTRODUCTION

We report on a Ti:sapphire laser system which produces 1 mJ pulses with a repetition rate of 1 kHz.[1] At this repetition rate, milijoule-level pulses represent a significant increase in energy for ultrashort pulses.[2-4] With a 21 fs pulse duration, our pulses achieve a peak power of 0.05 TW. The entire system including pump lasers can fit on about half of an optical table, and it is less expensive than its larger counterparts which produce higher pulse energies but at lower repetition rates.[5] In our laser, we have utilized a novel multipass ring design which reduces higher-order dispersion and preserves amplified bandwidth in comparison with a standard regenerative amplifier design, which employs an intracavity Pockels cell and polarizers.

We have used our 1 kHz laser system to investigate third harmonic conversion in a gas cell. The laser wavelength is centered at ~800 nm which when converted to the third harmonic gives a wavelength of ~267 nm. Harmonic generation in a gas rather than in a solid crystal has the advantage of a wider bandwidth acceptance for phase-matching, and hence the potential for much shorter pulse durations. The 1 mJ laser pulses are converted into the third harmonic with an efficiency of 0.1%, generating UV pulses of energy 1 μJ. At our 1 kHz repetition rate, this gives an average UV power of 1 mW. We have measured the duration of the UV pulses produced in air and found it to be ~16 fs.

2. LASER SYSTEM

The seed pulses used in our system originate in a Ti:sapphire oscillator which has been described in detail elsewhere.[6] The 4 nJ, 15 fs pulses from the oscillator are stretched to about 45 ps by an all reflective pulse stretcher, also described elsewhere.[5,7] A Pockels cell selects a seed pulse at 1 kHz, and the pulses are injected into a three-mirror ring multipass amplifier for a total of 8 passes. Fig. 1 shows a schematic of the laser system. The amplifier configuration consists of two identical focusing mirrors, focal length 50 cm, which are separated by the sum of their focal lengths and directed towards each other with slight horizontal misalignment. A flat mirror is situated midway between and perpendicular to the two mirrors and is approximately 14 cm away from the line intersecting them. Using this configuration, the beam makes 8 passes around the triangular loop, while on each pass reflecting from different points on the mirrors. Two additional mirrors inject and extract the beam after the desired number of passes. The multipass scheme alleviates the need for a Pockels cell or polarizers such as in a regenerative amplifier design and helps to avoid unwanted dispersion and spectral narrowing.

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The beam on each round trip focuses at the midpoint between the two curved mirrors, and this is where the amplifying medium is positioned. The injected seed pulse is initially collimated with a 2 mm diameter, and it focuses to ~ 300 μm in the crystal. At the crossing region the center of each passing beam is within 250 μm of any other. The eight beam paths are contained in a single horizontal plane. The first and final beams emerging from the crossing region form an angle <5 degrees. Fig. 2 shows a calculation of the beam propagation in the amplifier configuration where the beam on each pass is treated as a single ray. The top graph shows the entire setup; the middle graph shows a close-up view of the rays hitting the flat mirror region; the lower graph shows a close-up view of the crossing region where the amplifying crystal is positioned. The crossing region is seen to have a width of ~250 μm. This width grows when the number of passes is increased (under the constraint that the beam paths cannot overlap near the flat mirror where injection and extraction occurs). When mirrors of unequal focal lengths are used, it is not possible to get the beams to cross in a small region.

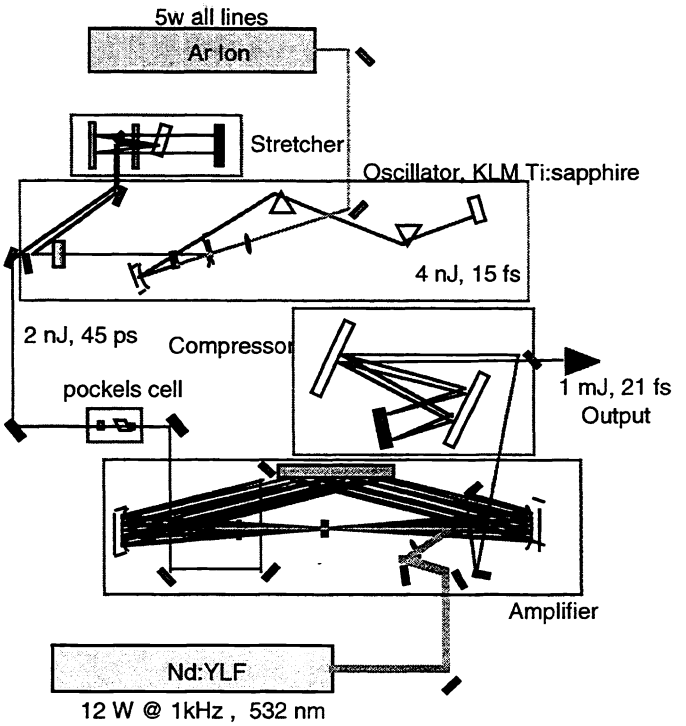
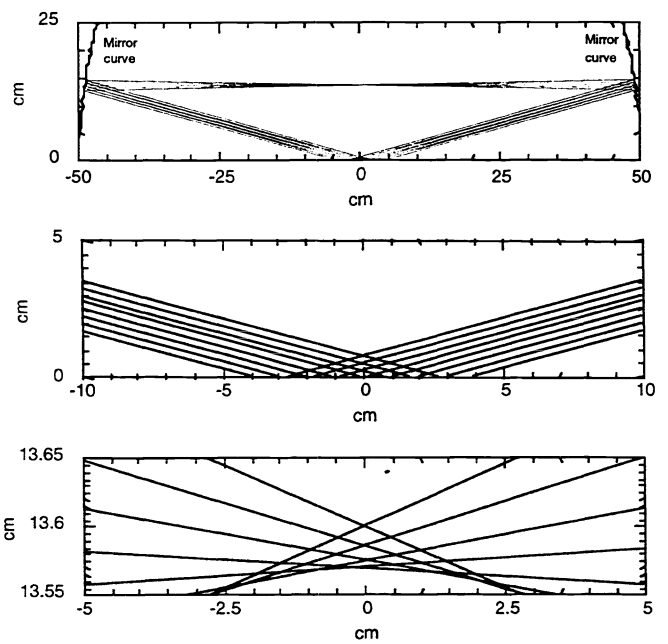


Figure 1. Schematic of laser system. The pulse originates in the oscillator and is then temporally stretched to 45 ps. The Pockels selects a single pulse to be injected into the amplifier ring each millisecond. A mirror picks off the beam after 8th pass through the amplifier, and the beam is then temporally compressed to 21 fs. The output energy is ~1 mJ per pulse.



The amplifier is pumped by 12 mJ pulses of 532 nm light at a repetition rate of 1 kHz. The pump beam is focused into an 8 mm Brewster-cut, Ti:Sapphire crystal with 0.2% Titanium doping. The pump spot size on the crystal is ~800 μm ($1/e^2$ diameter). On the first pass through the Ti:sapphire crystal, 74% of the pump light is absorbed. The residual energy is collected and refocused back into the crystal from the opposite side. To remove excess heat from the crystal, we circulate ethylene-glycol at ~ 0^{o} C through the copper mount holding the amplifier crystal.

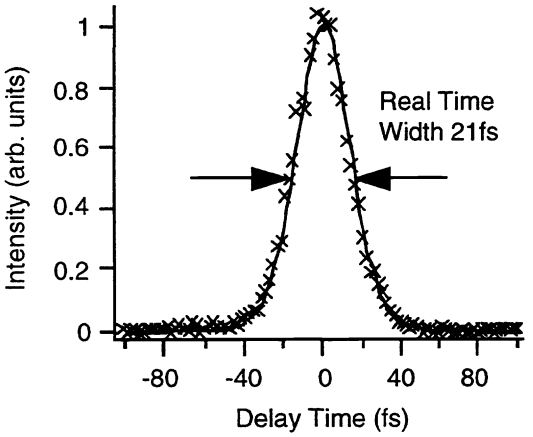
Figure 2 Modeling of beam propagation within the laser amplifier ring. The lower two plots give close-up views of different regions.

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A mask with separate 1 mm holes for each beam pass is placed near the flat mirror. The mask reduces Amplified Spontaneous Emission (ASE) and compensates for a thermal lensing effect due to high average pump power. If the beam in the amplifier ring is not restricted by the mask, the spot size on the curved mirrors grows with each successive pass. This results in a tighter spot in the crystal which extracts pump energy less efficiently, and, if left unchecked, can damage the crystal. After amplification, the pulses are temporally compressed by a double pass on a grating pair of the same type as described by J. Zhou et al.[5,7]

3. BEAM CHARACTERIZATION

For a pump pulse energy of 12 mJ , the output energy from the amplifier is 1.6 mJ. The pulses are able to be compressed to 21 fs while preserving 0.9 mJ of the energy. This is more than 2 times the output energy and less than half of the pulse duration produced by previously reported Ti:Sapphire system with a ~1 kHz repetition rate.[2-4]



A single-shot autocorrelator was used to measure the amplified pulse duration. An autocorrelation trace corresponding to a 21 ± 1 fs pulse is shown in Fig. 3. To determine the duration of the pulse, an autocorrelated $Sech^2$ function was fit to the data, and the full-width at half maximum of the laser pulse was extracted. The measurement suggests that the pulse is near the transform limit implied by the spectral width, 40 nm, which would imply a ~18 fs pulse. The beam exhibits a Gaussian-like spatial profile and focuses to 1.8 times the diffraction limit. More recently, we have characterized the pulse using the method of frequency-resolved optical gating (FROG)[8] and found good agreement with the autocorrelation measurement.

We were able to determine that ASE contributes at most 3% to the pulse energy. It appears that the injected pulse depletes the amount of gain available to ASE since ASE rises to about 25% of the pulse energy when the seed pulse is blocked.

Figure 3. An autocorrelation measurement and best fit of an autocorrelated $Sech^2$ function. The pulse energy was 0.8 mJ, and the fitted curve corresponds to a pulse with a full-width at half maximum of 21 fs.

4. THIRD HARMONIC CONVERSION IN GASES

Using our laser pulses, we investigated third harmonic conversion in gases, mainly air and argon.[9] Fig. 4 shows a diagram of the setup including the setup for diagnosing UV pulse durations. The laser pulses were focused with a 30 cm-focal-length mirror resulting f/30 focusing. At 1 mJ, this corresponds to a nominal peak intensity of ~10^{16} W/cm^2 if the focus is devoid of gas. Such a high intensity ensures a strong ionization break down of the gas, and hence, a radical distortion to the laser beam as it propagates through the focus. Therefore, it is unlikely that the intensity reaches this nominal intensity. With a pressure near an atmosphere, the laser produces a plasma channel ~1 cm long (indicated by the length of the recombination light).

The output light was collected and recollimated by a f = 25 cm dichroic mirror which reflected >95% of 267 nm light and transmitted >90% of the laser light. The pulse was further reflected from three additional mirrors having the same dichroic coating. The mirrors reduce the fundamental light by a factor >10^4 while negligibly attenuating the UV light. We verified that evacuating the focus caused the third harmonic signal to be extinguished.

As shown in Fig. 4, we used a standard self-diffraction FROG setup[8] to measure the pulse duration of the third harmonic light. In order to avoid temporal dispersion from propagation in a material, the beam is split by spatially cutting the beam in half as it hits the edge of a mirror. One beam is delayed relative to the other and they are brought back together near the focus of an f = 15 cm mirror where they cross with a 1-degree angle. A 100 μm thick sapphire plate is placed at the crossing point. The two crossing pulses diffract each other, owing to a spatially periodic index change induced by the Kerr effect.

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This causes faint self-diffracted beams to emerge outside of the two original beams at angles equal to their crossing angle. We monitored the spectral amplitude of one of the self-diffracted beams as a function of relative delay between the pulses. The self-diffraction FROG algorithm[8] reconstructed from the data a pulse duration of 16 fs for the third harmonic light generated in an atmosphere of air. Similarly short pulses were measured for third harmonics generated in Argon. The process of self diffraction as a means of mixing the two beams is convenient because it does not require the second harmonic conversion of the UV pulses, and thus avoids the problem of working with vacuum ultraviolet light.

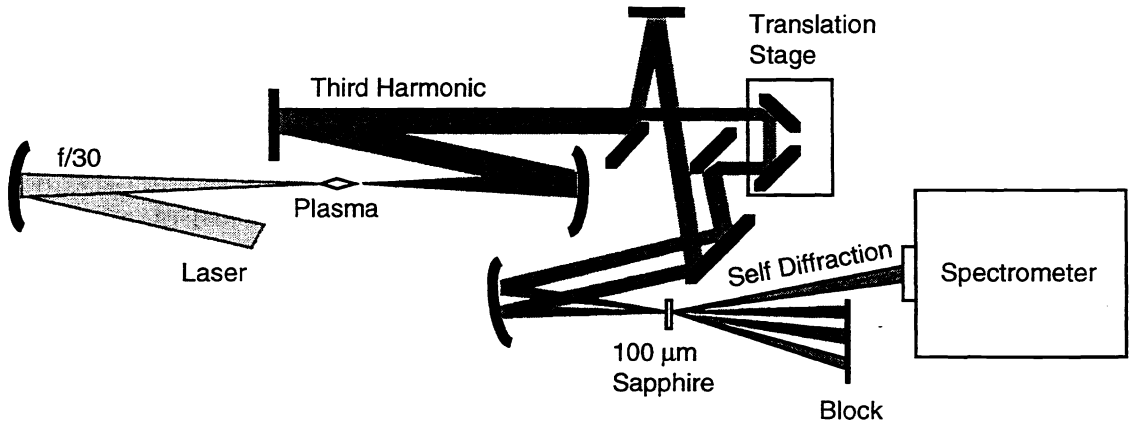


Figure 4. Setup for production and pulse duration measurement of third harmonic light. Four dichroic mirrors in the setup after the focus jointly attenuate the laser relative to the UV light by a factor $>10^4$.

Fig. 5 shows the measured third harmonic pulse energy as a function of argon gas pressure. The highest third harmonic energy, 1.1 μJ, occurs for a pressure of about half an atmosphere. (When we negatively chirp our laser pulse to about 40 fs, the UV pulse energy reaches 1.4 μJ.) This gives an efficiency of >0.1% for the conversion of laser light into the third harmonic. The result is similar for air, although with about 60% the conversion efficiency of argon. The measured bandwidth of the third harmonic produced in either air or argon is about ~6 nm. This can in principle support a pulse as short as 12 fs, rather than the 16 fs measured in air. The difference may be due to the dispersion of the air in the beam path of the FROG setup.

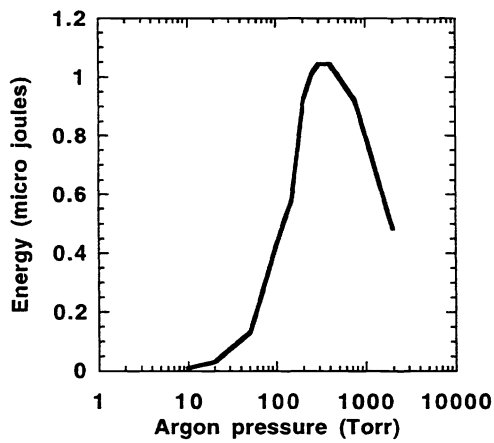


Figure 5. Third harmonic pulse energy as a function of argon pressure.

Our work on third harmonic production is similar to that of Siders et al.[10] who reported efficiencies of 0.01% for third harmonic conversion in argon of their 80 fs, 1.5 mJ Ti:sapphire laser focused using f/10 optics. Blueshifting allowed them to achieve UV bandwidths as wide as those of our UV pulses, but they did not measure the actual pulse duration. We investigated third harmonic production using f/10 optics with our laser, but we found that the conversion efficiency diminished to a level similar to that seen by Siders et al. Under such conditions, the fundamental light is dramatically blue-shifted (many visible colors) and scattered into a cone angle much wider than that of the initial f-number. For our laser conditions, the highest third harmonic output occurs for f/30 focusing, and this results in a conversion efficiency ten times

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higher than had been seen previously. At significantly larger f-numbers the third harmonic emission decreases, and the blue shifting and scattering of the laser pulse is no longer apparent.

5. SPATIAL AND SPECTRAL CHARACTERIZATION OF FUNDAMENTAL AND THIRD HARMONIC BEAMS

When the intense laser pulse is focused into the gaseous medium, the propagation of the beam and hence, the nature of the focusing is affected. The electronic nonlinearities of the neutral atoms and the temporally and spatially inhomogeneous appearance of free electrons through ionization causes self-phase modulation, defocusing, and blueshifting of the light. To gain insight into this complex interaction we conducted a study of the spectral and spatial properties of the emerging light. We obtained two-dimensional maps of the emerging light where the spectral content of the light was plotted against the radial position in the beam.[11] These maps are of the same type calculated by S. C. Rae,[12] although obtained for very different laser conditions.

The quality of the laser spatial profile is of particular importance to this measurement because small distortions in the laser profile can translate into severe asymmetries in the spatial structure of the radiation emerging from the interaction region. When we focus our 1.8-times diffraction limited beam into the gas, the fundamental and third harmonic light beams emerge with complicated and azimuthally asymmetric spatial patterns. This prevents a meaningful interpretation of the spatial structure of the emerging light since the laser beam in principle has azimuthal symmetry. In order to produce a near-perfect incident beam, we spatially filtered the beam of our 10 Hz, 25 fs, 2 TW Ti:Sapphire system,[5] at the expense of nearly all of the pulse energy. This resulted in a beam ~1.05 times the diffraction limit with an energy comparable to the 1 kHz system described in this paper.

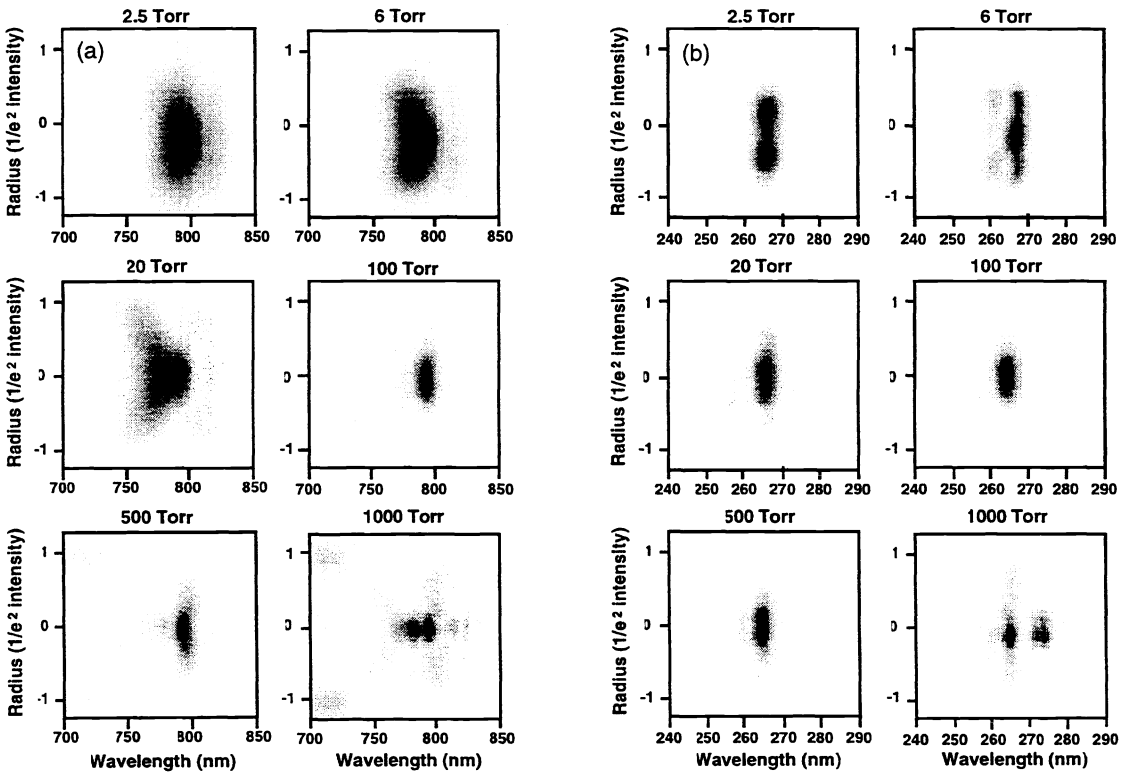


Figure 6. The (a) fundamental and (b) third harmonic spectra as a function of beam radial position for a pulse energy of 0.9 mJ focused in various pressures of argon. Position is expressed in units of $1/e^2$-intensity radius of the beam when the gas cell is evacuated. The brightness of the images should not be compared from frame to frame.

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Fig. 6 shows spectral-spatial maps of the emerging light well after the focus, recorded for various pressures of argon. In all cases, the maps show good azimuthal symmetry, indicating that the spatial filtering of the laser beam is sufficient to make the measurement meaningful. The maps were made for both the (a) fundamental and (b) third harmonic light. The measurements indicate that the optimal spatial quality of the third-harmonic beam occurs at pressures in the range of ~300 Torr, approximately the same pressure where the highest conversion efficiency takes place (see Fig. 5). They also show that at the highest pressures, the fundamental and UV beams undergo severe distortions.

6. SUMMARY

We have constructed a Ti:sapphire amplifier system which produces 21 fs, 1 mJ pulses at 1 kHz. The amplifier crystal is pumped with 12 mJ of 527 nm light which is converted into amplified pulse energy with an efficiency exceeding 10%. The beam focuses to ~1.8 times the diffraction limit. We have used the laser to produce third harmonic light in a gas cell, obtaining a conversion efficiency above 0.1%. We found that for our laser conditions, the optimal conversion efficiency is obtained for f/30 optics at gas pressures near one half atmosphere. The duration of the 267 nm, 1 μJ pulses was measured to be ~16 fs, representing the shortest UV pulses measured to date. We have recorded spectral-spatial maps of the third harmonic light emerging from the focus for various pressures of argon, where the spatial quality of the incident laser beam is near the diffraction limit. We find that the spectral and spatial characteristics of the third harmonic beam are well behaved for the range of pressure where the highest conversion efficiency occurs. The process of focusing an intense laser into a gas cell is complicated, and these measurements may be useful to compare with numerical simulations which often employ simplifying approximations. We are working on such calculations and plan to report on them in the future.

7. ACKNOWLEDGMENT

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