Novel laser system for ultrafast spectroscopy

Matthew A. Horn\textsuperscript{a}, David C. Arnett\textsuperscript{b}, Norbert F. Scherer\textsuperscript{a}

\textsuperscript{a}Department of Chemistry and James Franck Institute, University of Chicago, Chicago, IL 60637
\textsuperscript{b}Pacific Northwest National Lab, Richland, WA 99352

ABSTRACT

The design and performance of a cavity-dumped Ti:sapphire (CD-TS) oscillator and short pulse-pumped amplifier system is presented. Dumped pulses as short as 12 fs duration and 60 nJ in energy, with 70\%-80\% dumping efficiency are made possible by a stretched dumper-arm configuration. The oscillator yields stable, reliable, and reproducible pulses. Pulse duration and phase structure are analyzed via wavelength-resolved nonresonant (solvent) scattering (WRNS). Single and multi-pass amplification schemes and performance results are described. The optical continuum generated using single-pass amplified pulses is characterized by frequency-resolved cross-correlations.

1. INTRODUCTION

Ultrafast condensed phase spectroscopy is a field which demands intense and very short pulses of light over a broad range of frequencies. Coherence studies, including those of the photosynthetic reaction center, can profitably utilize pulses less than 10 fs in duration since solute-solvent interactions can occur on the sub-10 fs timescale.\textsuperscript{1,2,3} Terahertz spectroscopy requires intense laser fields both to produce and to detect the terahertz emissions.\textsuperscript{4} Nonlinear optical spectroscopies in general, including EFISH interfacial studies, benefit from intense laser fields to produce the, for example second harmonic, signal.\textsuperscript{5} All these studies, while widely disparate in content, require a laser system capable of providing intense, short duration pulses.

The past twenty years have seen tremendous advances in the preparation of such pulses. In the 1980s mode-locked dye oscillators and low repetition rate dye amplifiers gave spectroscopists access to the 100 fs regime.\textsuperscript{6} Shank and co-workers took this technology to its extreme with 6 fs pulses.\textsuperscript{7} In the 1990s Kerr-lens mode-locked (KLM) Ti:sapphire oscillators and amplifiers became the system of choice in many laboratories. As this decade is rounding out, conventional Ti:sapphire oscillators have produced pulses of less than 10 fs duration and Ti:sapphire oscillator and amplifier systems have produced pulses of less than 30 fs.\textsuperscript{8} Within the last year the development of chirped mirrors and continuum generation and compression have pushed the state-of-the-art to the point of pulses less than 5 fs in duration.\textsuperscript{9,10}

These advances in pulse length, along with an analogous advance in pulse intensity, have facilitated the study of many phenomena, but at the cost of system simplicity. Bragg cells, amplifiers, stretchers, and fibers (or other continuum generators) all add dispersion to the pulses that compressors and dispersion compensation lines attempt to remove. Both add experimental difficulty and cost. A desirable advance would be a spectroscopic system that is able to provide short and intense pulses while also providing a minimum of difficulty. The system detailed here was designed with that desirable advance in mind. It entails a cavity-dumped Ti:sapphire (CD-TS) oscillator, a simple short-pulse pumped multi-pass amplifier, a stretcher and compressor and a continuum generator. In addition to describing the laser system design, characterization of the continuum and utility for pump-probe measurements will be discussed.

2. OSCILLATOR

The CD-TS oscillator design presented here improves dumping efficiency while simultaneously decreasing sensitivity to alignment. The oscillator is a modified Murnane designed oscillator, pumped with an argon-ion laser (Coherent 310) or a frequency-doubled Nd:YVO\textsubscript{4} laser (Spectra-Physics Millennia). The unmodified oscillator is a standard four-mirror cavity, with two 10 cm radius of curvature mirrors focussing into the Ti:sapphire and fused silica prisms spaced 75 cm apart. The modification is the addition of a stretched cavity-dumper arm pictured in Figure 1. The additional fold for the Bragg cell is at an 11° angle to compensate for the additional astigmatism.\textsuperscript{11} The Bragg cell is placed at the confocal focus of mirrors M3 and M4. A low-percentage output coupler is placed in the prism arm rather than the cavity-dumper arm for this cavity. The high-reflecting end mirror is placed 36 cm away from the Bragg cell.
This 36 cm distance is a unique feature of this oscillator. Previous cavity-dumped lasers were designed to minimize the temporal delay between the forward and rearward interactions in the double-pass Bragg cell arrangement; this required short radius of curvature mirrors and very tight focusing conditions, both of which introduce instability into the laser. The present design does not take this approach and instead, as shown in Figure 2, has the optical pulse interact with consecutive maxima of the oscillatory rf waveform driving the Bragg cell. This allows the use of looser focusing conditions (greater radius of curvature mirrors) thereby increasing the stability of the laser for pulsed operation and making the laser less sensitive to the alignment of the system. An additional advantage of this approach is that the efficiency of dumping can be higher, because both forward and rearward interactions occur at the maximum of an rf pulse oscillation; previous approaches dictated that either forward or rearward diffraction happens off of a maximum. Dumping efficiency of 80% has been achieved in the three CD-TS lasers that have been in operation for three years in the Scherer group.

Figure 2. Cavity dumping approach. P: pick-off mirror; M3,M4: 15 cm roc mirrors; B: Bragg cell; EM: end mirror.

Operationally, cavity-dumping is performed by driving the Bragg cell with a synchronized rf source (CAMAC CD-5000). Synchronization with the 76 MHz cavity repetition rate is achieved with a fast photodiode that samples the beam transmitted through the output coupler. This beam passes through an adjustable neutral density filter and is focussed onto the photodiode. The level of light is adjusted to optimize the signal sent to the Bragg cell driver. This signal acts as the master clock for the entire system. Additional timing control electronics are added to synchronize the dumping with other devices to be described later. The timing box (CAMAC TS 2004 Timing System) also allows the Bragg cell driver to function at lower dumping rates (below 10 kHz). If the rf waveform is passed through an 18 Watt rf amplifier (CAMAC PB 1800) a dumping efficiency of 80% can be achieved that is stable for an entire workday. (Higher dumping efficiencies can be achieved, but at the risk of extinguishing pulsing.) Without the rf amplifier, good stability can be achieved for efficiencies up to 70%. Typical pulse energies range from 40-60 nJ per pulse when the cavity is arranged for short pulse production—12-15 fs duration pulses. Pulses with energies in excess of 80 nJ have been obtained, but temporal broadening occurs in this high-power configuration. Optical continuum generation in single mode optical fibers is achievable as also reported by others.10
The intracavity energy recovers to a maximum approximately 500 ns after dumping. This recovery time sets an upper limit for the rate of cavity dumping (~1 MHz). The intracavity energy exhibits relaxation oscillations as it decays to the steady-state level in 7 \( \mu s \). All of the energy is contained in one pulse—pre-pulse to main pulse ratios in the present CD-TS system are typically on the order of three hundred to one, and can be tuned by shortening the rf pulse duration to be as high as 1000:1. Typical shot-to-shot noise in the dumped pulse train is much less than one percent. The cavity-dumping technique introduces an insignificant amount of noise and the oscillator is nearly as stable as the conventional four-mirror oscillator.

### 3. PULSE CHARACTERIZATION

Many non-linear spectroscopy experiments require a precise determination of the temporal (i.e. pump-probe) delay between optical pulses, and at least a nominal description of the temporal pulse-width and pulse phase structure.\(^\text{13}\) This information can be garnered from a number of measurements, including autocorrelations, cross-correlations, and a whole variety of frequency-resolved optical gating techniques (FROG).\(^\text{14}\) From an experimental viewpoint, it is preferable to obtain all of this information from a measurement done “in situ” through a technique termed wavelength resolved nonresonant scattering (WRNS),\(^\text{1}\) which is essentially the transient grating FROG technique (TG-FROG) discussed recently by Trebino and co-workers.\(^\text{15}\)

Following compensation for positive group velocity dispersion (GVD), which originates from the amplified CD-TS system and several beam-splitters and waveplates, the pulse train is split into three beams of approximately equal intensity for measurements such as photon echo or transient grating. The three beams, with momentum vectors \( k_1, k_2, \) and \( k_3 \) are sent through variable delay lines and focussed into the sample using an all-reflective Cassegrain telescope (\( M1 = 40 \text{ cm}, M2 = -100 \text{ cm} \)) to a spot of less than 200 microns diameter at a crossing angle of less than two degrees. Loose focussing conditions and a small crossing angle are necessary to avoid excitation saturation, sample degradation, or spurious photon echo results for many samples. Tighter focussing conditions have been employed for hardier samples. For all the measurements presented here, the three excitation beams are arranged in the boxcar geometry. The signal appears on the fourth corner of a square defined by the three excitation beams, in the phase and momentum matched direction \( k_s = k_3 + k_2 - k_1 \). The scattering signal is dispersed in a monochromator and detected with an avalanche photodiode (Hamamatsu).

The TG-FROG technique was recently demonstrated for 1 \( \mu J \) pulses at 426 nm.\(^\text{15}\) In this case, three excitation beams in a boxcars geometry were allowed to interact in the non-linear medium of fused silica. The pulses generated by the CD-TS laser described here have substantially less energy than the pulses used by Trebino and co-workers, and so the signal level benefits from having a medium with larger non-linearity to generate an easily measurable TG-FROG type signal. In highly polarizable liquids such as CS\(_2\) or pyridine, the non-resonant electronic response of the liquid provides a dominant contribution to the scattered signal which can be wavelength-resolved to generate a WRNS trace. This technique is particularly advantageous for solution phase studies, allowing for complete characterization of the instrument response function through the measurement of a “solvent blank.”

The WRNS result for cavity-dumped pulses is shown in Figure 3. A solvent response of 18 fs FWHM is measured with no wavelength-dependent change in temporal width or a shift in the zero of time indicating that linear chirp has been accounted for. The spectrum of the cavity-dumped pulses is about 100 nm in width and centered at 800 nm. Such cavity-dumped pulses have been used for photon echo experiments in this group\(^\text{16}\) and others.\(^\text{17,18}\) The controlled repetition rate and high peak power facilitates examining air-sensitive samples in spinning cells and reaction dynamics of molecules with small extinction coefficients\(^\text{19}\) and in situ WRNS pulse characterization.
4. SINGLE-PASS AMPLIFICATION

While a CD-TS system is appropriate for studying many chemical and material systems, many applications remain where higher pulse energies are desirable. Amplification is necessary to achieve this. Regenerative amplifiers impart a great deal of dispersion (linear and higher order) to the optical pulse and thus the amplified pulses cannot be compressed to their original short length.\textsuperscript{20} A more satisfactory amplification scheme is one that is simple to use and imparts little dispersion to the pulse. Multi-pass amplification schemes have become more common recently.\textsuperscript{21} However, multi-pass schemes using a long-duration, high-energy pump pulse are not very efficient in gain extraction since the pump and Ti:sapphire beams are not collinear, as is the case with regenerative amplifiers. An amplifier that combines the best aspects of these two designs is desired.

Two such designs have been employed, single-pass, pictured in Figure 4, and double-pass, pictured in Figure 8. Both are pumped by a Q-switched frequency-doubled Nd:YAG laser. A home-built (pumped by a 10 Watt (SDL) diode laser) Nd:YAG laser was employed for the single-pass amplifier and a Spectra-Physics T40 system for the double-pass amplifier. The home-built amplifier pump laser is a short (20 cm) linear cavity with a 10 mm Nd:YAG rod and an intra-cavity Q-switch. The Nd:YAG rod is coated such that it is also an end mirror for the cavity and is longitudinally pumped by the fiber-coupled diode laser output. The thin film polarizer was inserted into the cavity to improve mode quality, pulse stability and holdoff. The frequency doubling was performed by focussing the 1064 nm light into a 10 mm NCPM type II LBO crystal. In both systems the Q-switches are synchronized with the pulses from the CD-TS oscillator via the electronic timing system as shown in Figure 4. The home-built system produces 100 µJ and the commercial system 0.4 millijoule pulses at 532 nm at 4 kHz repetition rate. Both systems passed the green pump beam through a λ/2 waveplate before focussing and mode matching the beam into the amplifier Ti:sapphire slab. Both systems employ a simple x-fold configuration with 10 cm radius of curvature mirrors reflecting the near IR beams at 11° to minimize astigmatism.\textsuperscript{11} Focussing conditions are tight to ensure efficient extraction. Both systems are carefully mode matched throughout the entire beam path.

The single-pass system has the two beams (Ti:sapphire and frequency-doubled Nd:YAG) counterpropagate to maximize energy extraction. In this scheme the pulses are routed directly from the cavity to the amplifier without stretching. Proper alignment yields a gain factor of approximately twenty; this produces pulses low enough in energy (0.5 – 1 µJ per pulse) that material damage of the Ti:sapphire gain medium is not a problem. This amplification scheme adopts many of the strong points associated with regenerative and multi-pass amplification and provides pulse energies suitable for many nonlinear spectroscopic applications. The incorporation of only a single pass limits the amount of dispersive material introduced into the optical beam. These considerations render stretchers and compressors unnecessary and allow for dispersion compensation with a simple pair of fused silica prisms. The absence of spectrally narrowing optics (thin film polarizers, waveplates) and highly dispersive electro-optic or acousto-optic isolation devices allows the broad CD-TS spectrum to be maintained throughout the amplification process.
Figure 4. Single-pass amplification scheme. A1, A2: 20 cm roc mirrors; OC: output coupler; λ/2: half-wave plate; λ/4: quarter-wave plate; Y: 10 mm Nd:YAG rod; QS: Q-switch; TFP: thin film polarizer; TAMP: Ti:sapphire.

The laser spectra are comparable prior to and after amplification, as shown in Figure 5, exhibiting only slight spectral narrowing as a result of a slight mismatch in the spectral properties of the amplifier curved mirrors used to generate this spectrum. The amplified pulse spectrum still has a spectral width of 90 nm. When the mirrors are carefully matched to the spectrum of the laser, no spectral narrowing is observed. Autocorrelation techniques indicate that the CD-TS pulsewidth is completely recoverable after amplification and dispersion compensation with a prism pair.

Figure 5. Unamplified and amplified laser spectra.

The single-pass amplification system was used to produce a continuum by focusing the pulses into a sapphire disc. These continuum pulses were analyzed by a cross-correlation technique. A small portion (~10%) of the amplified pulses were reflected off of a window before the remainder was focused into the disc. The picked off portion was passed through a sapphire disc to match the dispersion imparted to both beams. Both beams were then focused into a KDP crystal, and subsequently the sum frequency signal was dispersed in a monochromator and these wavelength-resolved cross-correlations measured. One of these, the component at 7.5×10^{14} Hz (i.e. 400 nm), can be seen in Figure 6. The FWHM of the cross-correlation is 28 fs; considering that the incident pulse width is about 15 fs this yields a continuum (gaussian) pulse of 23 fs duration.
Figure 6. Wavelength-resolved cross-correlation of continuum. Sum frequency detection is at 400 nm.

The pulsewidth and temporal peak shift of each frequency component is presented in Figure 7. Short duration continuum pulses of 10 fs (gaussian) are obtained around the 425 nm detection window. Clearly the dispersion imparted to the pulses is not linear in this set-up; a prism pair does not optimally re-compress these pulses across the entire spectral range. In particular, significant broadening does occur over the blue end of the spectrum. The solid line through the peak position data indicates a cubic chirp (curvature) in addition to GVD.

Figure 7. Peak shift and width of continuum cross-correlations versus frequency. The solid lines through the two data sets are polynomial least squares best fits. The polynomial equation shown as an inset refers to the Peak Position versus Frequency data.

5. DOUBLE-PASS AMPLIFICATION

In spite of the advantages of the single-pass technique some applications remain where higher powers are desirable. Also, increased stability in amplification occurs under gain and extraction saturation conditions. The two-pass amplification gives this extra power and moves further toward saturation; the scheme is shown in Figure 8. The peak powers, however, for unstretched pulses are now above the threshold for material damage, thus stretchers and compressors are now necessary. Their designs are described below. Also, the double-pass amplification technique sends the beam backward along its original trajectory and thus a uni-directional device must also be incorporated. A unique low dispersion device is also described below.
The retroreflection of the double-pass amplification scheme demands a uni-directional device to extract the amplified beam from the oscillator-amplifier system before the pulse re-enters the oscillator cavity. Many systems utilize a Pockels cell to rotate the polarization of the pulse in the amplifier and then reflect the altered pulse off of a polarization-sensitive reflector. In a regenerative amplification arrangement this commonly means that a large amount of dispersion is being added to the pulse as it passes through long electro-optic crystals and Faraday rotators many times.

The solution to this problem presented here is firstly to limit the number of passes to two such that less material dispersion is added. Still the amplification can be as high as a factor of 400, given a single-pass gain factor of twenty. Resultant pulse energies are into the ten microjoule regime—energies high enough for most nonlinear optical processes. Secondly, the use of polarization optics to extract the pulse were altogether avoided thus eliminating the inefficiencies of polarization-sensitive reflectors and bulky Pockels cells. Instead, the present system utilizes an acousto-optic (AO) modulator as a uni-directional device (IntraAction AOM 125 driven with IntraAction ME signal processor). It is synchronized with electronics to the same master clock as the rest of the system (76MHz) via the CAMAC timing system. In the forward direction the AO modulator deflects 50% of the beam into the amplifier. The AO modulator “turns off” before the rearward pulse returns to the modulator. Thus the retroreflecting amplified pulses pass through undeflected and in a new direction from the original beam path. This new, amplified beam can then be picked off and sent to the compressor.

Pulse stretchers and compressors are usually difficult elements to align and often have low throughput. The stretcher and compressor employed here are based on the design of Wynne and Hochstrasser and pictured in Figure 9. The advantage of this design is its simplicity. Both stretcher and compressor use identical elements: a single diffraction grating, a spherical mirror, and three flat mirrors. The input beam hits mirror M1 and reflects off of the grating (Milton Roy, 300 grooves/mm) at the Littrow angle. The retro-reflected beam passes just over M1 and onto the spherical mirror M2. This retro-reflected beam passes just over the grating onto M3, which is positioned in the focal plane of M2. M3 also retroreflects the beam, sending it back to M2 just below its original position. This beam returns to the grating (again at Littrow angle) and is reflected onto M4, which sends the beam back on itself with a small vertical displacement so that the beam can be picked off outside of the stretcher “cavity”. Small vertical offsets ensure no spatial dispersion. The mirrors M2 and M3 form a telescope with unit magnification, and thus if the grating is placed before the telescope (i.e. between M1 and M2) the grating introduces positive group-velocity dispersion (GVD); conversely, if the grating comes after M3 it introduces negative GVD. Thus, a stretcher and compressor pair can be made to match simply by matching the distance of the grating from the telescope. For optimum femtosecond pulse durations this distance must be well-matched to within a few micrometers. Compensating for the small dispersion encountered in the amplifier is also simply achieved by translating the grating. The angles are matched between the stretcher and compressor by fixing the positions in the stretcher and using high-quality rotators and translators in the compressor to match the stretcher. The angles of the gratings must match to within 0.1°.
6. FUTURE DIRECTIONS

Current work is ongoing to extend this system to a gain-switched several pass regenerative amplifier design. Studies are underway utilizing these low dispersion amplified pulses in optical pump-continuum probe experiments and OPA pumping.

7. ACKNOWLEDGMENTS

This work was supported by a grant from the National Science Foundation (CHE93-57424), and fellowships from the David and Lucille Packard, Camille Dreyfus, and Alfred P. Sloan Foundations.

8. REFERENCES


