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MICHIGAN TO LAWRENCE LIVERMORE
NATIONAL LABORATORY

ATTENTION OF:

Mr. H.A. PITKIN
Mail Stop L650
Lawrence Livermore National Laboratory
PO BOX 5012
Livermore, California 94551

5012 1991

PRINCIPAL INVESTIGATORSP:

Professor Gérard Mourou
and Dr. Philippe Bado
University of Michigan
USL, Room 1006
2200 Bonisteel Blvd.
Ann Arbor, Michigan, 48109-2099

TITLE:

"Generation of High-Quality Petawatt
Pulses"

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II. DEVELOPMENT OF OSCILLATORS OPTIMIZED FOR CHIRPED PULSE AMPLIFICATION.

CPA sources need a front-end capable of generating very short seed pulses. To meet this requirement, present CPA sources rely on actively modelocked solid-state oscillators used in conjunction with non-linear fiber stages. This approach generates pulses with a residual frequency chirp, resulting in a limited peak-to-background intensity contrast ratio. The availability of an oscillator capable producing directly (i.e. without fiber stage) picosecond or sub-picosecond pulses would significantly improve the quality of CPA sources.

We have pursued different approaches to replace the standard modelocked oscillator front-end. In an initial phase, we investigated the possible use of Additive Pulse Modelocking (APM) color-center lasers as oscillators for CPA Nd:glass sources. Lately we have developed a Ti:Sapphire modelocked oscillator operating in the one-micron range.

In order to generate pulses with very high peak to background contrast ratio, we have built a non-linear coupled-cavity oscillator generating 200-fs pulses. This color-center laser operates in the 1.5-1.6 micron range. A diagram of the oscillator is shown in Fig. 1. The laser is pumped with a modelocked Nd:YAG source (3.7 W @ 1.06 μm). Both KCl and NaCl crystals were tested as gain media. The respective tuning curves are shown in Fig. 2.

NaCl was found to have a larger tuning range and to generate a higher average power (up to 150 mW). When synchronously mode-locked, the color-center generates pulses in the 10-20 ps range. Once interferometrically coupled to non-linear external-cavity, this oscillator produces very short pulses (85 to 260 fs). This oscillator was first operated with a 2 mm birefringent plate. 85 fs, near transform-limited pulses were generated at 1.54 micron. As the laser was tuned to longer wavelength, the pulse duration increased, as well as the bandwidth-duration product. With a 4 mm birefringent plate, 125 fs, transform-limited pulses were generated over the full tuning range.

CPA oscillators must meet some very stringent stability requirements. APM lasers do require some type of feedback loop for stable, long-term operation. We have duplicated the feedback system originally proposed by Mollenauer.³ After addition of the feedback loop, we found that the oscillator could operate in numerous regimes, as predicted theoretically by Piché.⁴ Our experience shows that, at present, APM oscillators are not stable enough to be used at the front-end of a CPA chain.

Consequently, we have shifted our development emphasis to Ti:Sapphire based oscillators.

We have successfully demonstrated a stable modelocked oscillator operating in the one-micron range. The large tuning range (680-1100 nm) of Ti:sapphire indicates that it may be capable of generating picosecond, and sub-picosecond pulses. This broad bandwidth was first exploited through active mode-locking techniques,^{5,6,7} and has recently generated pulses as short as 1.3 ps.⁸ The use of a nonlinear external cavity based on the original work done by Mollenauer and Stolen⁹ on soliton lasers has also been adapted for use in the positive dispersion regime¹⁰ and was first employed with Ti:Sapphire by French and co-workers.¹¹ The various aspects of the mode-locking mechanisms and behavior of lasers which employ nonlinear external cavities has been developed by several groups.^{9,12,13,14} The first self-starting, cw mode-locked Ti:sapphire oscillator employing a nonlinear external cavity produced 1.4 ps pulses and through external dispersion compensation, 200 fs pulses.¹⁵ More recently, Spence et. al.¹⁶ were able to demonstrate a self-modelocked Ti:sapphire laser with internal dispersion compensation which produced femtosecond pulses without the use of an external cavity.

In each of the above cases the mode-locking behavior of the laser was examined at or near the 700-nm peak gain of Ti:sapphire (See Fig.3). We have recently mode-locked our Ti:sapphire laser in the 1 μ m wavelength regime using conventional active mode-locking techniques. We have been able to produce pulses as short as 1.7 ps directly from the oscillator. Passing these pulses through an extracavity grating pair arranged to provide negative GVD resulted in a further reduction of the pulse duration. Using this configuration pulses as short as 850 fs have been obtained.

The basic laser, a Spectra Physics model 3900,¹⁷ has been slightly modified for mode-locking operation and is shown in Fig. 4. The 1.8 m long, astigmatically compensated cavity is mounted on super-invar breadboard for optimum stability against thermal transients. A 3% output coupler is used, and mounted on a translation stage for fine cavity length adjustment. A two plate birefringent filter tunes the laser wavelength. The acousto-optics mode-locker (Spectra-Physics Model 451), is driven at 41 MHz, and is placed as close to the cavity end mirror as is physically possible.

Pumping with all lines of a cw argon-ion laser, the cavity lasing threshold (without mode-locker) is approximately 11 Watts. With mode-locker in place, the pump power threshold is raised to 16 Watts, with stable operation being achieved at 18-19 Watts. Average power out is 400 mW at 1.05 μ m (@19 Watts pump power). We have observed an unusual bistable behavior. The laser can start in two different and very stable regimes: a long pulse regime (50-60 ps) or a short pulse regime (2-3 ps). Mechanically perturbing one of the cavity mirrors transposes the laser from the long pulse to the short pulse regime. Pulse durations are typically 3-4 ps. Through careful optimization, we have achieved

pulses of 1.7 ps duration (a Gaussian profile is assumed since it is the usual assumption for actively mode-locked lasers). We have also attempted turning the mode-locker off to achieve the self-mode-locking as described by Spence.¹⁵ While we observe this behavior quite readily at 800 nm, we have not been able to achieve mode-locking in this manner at 1 μm .

Analysis of the pulse spectrum reveals that the pulses are far from transform limited. The typical spectrum width is 4.5-6.0 nm, depending on cavity alignment. The pulses are 10 times transform limited. The spectrum shape is most likely complicated by self-phase modulation occurring in the rod. This intricate shape indicates that the actual pulse profile may be far from Gaussian.

Some residual linear chirp could be removed with an extracavity grating pair (1200 l/mm, spaced 9 mm apart and at an angle slightly greater than Littrow) to produce 850 fs. Note that we cannot compress the pulse to the transform limit due to the complex initial phase law. Autocorrelation measurements carried over four orders of magnitude show a secondary pulse attributed to a reflection from one of the mode-locker surfaces. (The delay between the main and secondary pulse is found to depend upon the distance between the rear mirror and the modelocker). The energy ratio between the reflection and the main pulse is $\sim 10^{-3}$. This intensity contrast ratio is considerably better than for the standard Nd:YAG or Nd:YLF compressed pulses.

The tunability of the laser in terms of pulse duration and power out across the wavelength range 950-1070 nm (limit imposed by the mirror set), has also been examined. We have found that the pulse duration can be maintained throughout the wavelength range supported by the mirrors; while the output power is seen to increase as the wavelength is tuned toward 900 nm, as shown in Fig. 5.

The Kuizenga-Siegman mode-locking theory¹⁸ predicts a pulse duration longer than what has been measured experimentally. There are several possible reasons for this disagreement. First, the gain profile can no longer be assumed Gaussian. Our laser is tuned to a point in the gain profile where this assumption simply does not hold. Second, the intracavity intensities are high enough that significant nonlinear effects can occur within the Ti:sapphire rod. The magnitude of the nonlinear dephasing can be easily estimated. For 3% output coupling, and 400 mW power out, the intracavity average power is 13.3 Watts. For a 3 ps pulse in an 82 MHz wavetrain, we have an intracavity peak power of 54 kW. The beam waist at the rod is estimated to be 30 mm in diameter. This produces an intensity of 6 GW/cm². The Rayleigh range for a 30 mm spot at 1.054 μm is .13 cm, and will be used as the nonlinear interaction length. Using $n_2 = 3.1 \times 10^{-16} \text{ cm}^2/\text{W}$ as the nonlinear index of Ti:sapphire, we see that the nonlinear dephasing is around 4×10^{-2}

rad/pass. This value is of the order of the nonlinear dephasing obtained in passively mode-locked dye lasers,¹⁹ or in coupled-cavity lasers,¹⁴ and could explain the strong phase modulation observed, but is not sufficient to support a 2-ps pulse.²⁰

The rod birefringence may also play a key role in determining the pulse development.²¹ If the Ti:sapphire rod c-axis is not exactly aligned with the cavity polarization, the rod acts as a birefringent plate. When the nonlinearity due to self- and cross-phase modulation on each axis of the rod is included, the rod is actually an intensity dependent waveplate. This waveplate, along with the Brewsters' surfaces which act as polarizers, leads to a coupled-cavity type phenomenon. The two "cavities" are now the s and p polarizations. The common interface of the two "cavities" is any polarizer, or in this case, the Brewster angled surface of the rod. We are presently investigating this hypothesis²² which could aid in the production of pulses shorter than the limit predicted by the Kuizenga-Siegman theory for active mode-locking. Note however, that the dispersion between the polarizations limits the pulsewidth to approximately 700 fs. It is unlikely that this mechanism is responsible for producing the femtosecond pulses reported for other mode-locked Ti:Sapphire lasers.¹⁵

III. SPECTRAL NARROWING.

Recent application of the chirped pulse amplification (CPA) technique to the optical domain has made it possible to exploit the storage capability of solid-state laser media with ultra-short pulses. For instance, using Nd:glass amplifiers, 1 ps pulses have been amplified to the multi-joule level leading to peak powers in the 1-20 TW range. For these CPA sources, the final pulse duration is limited by spectral narrowing during the amplification process.²³ A careful examination reveals that most spectral narrowing occurs in the regenerative amplifier that is always present in CPA lasers. Typically, the pulse experiences a net energy gain of 10^8 - 10^9 through the regenerative amplifier (with a total small-signal gain around 10^{16}). Beyond the regenerative amplifier, the pulse is amplified by an additional factor of 10^3 - 10^4 (associated with a total small-signal gain of approximately 10^5). Thus, to significantly increase the final pulse bandwidth, the spectral narrowing collected in the regenerative amplifier must be eliminated. Typical spectra of the pulses before and after our Nd:glass regenerative amplifier are shown in Fig.6. A 3-fold reduction of the pulse spectrum width is typically observed (FWHM input spectrum: 4 nm, and an output energy of 3 mJ).

In order to reduce the gain narrowing present in the amplification chain, we have developed a simulation package analyzing the regenerative amplifier. Numerous cavity designs have been studied. Excellent agreement between theoretical and experimental data is obtained.

We have studied the possibility to broaden the amplifier gain bandwidth by using in the regenerative amplifier a combination of several types of Nd:glass. These glass were selected to offer a large combined bandwidth. The gain narrowing can be considerably reduced. Our theoretical study shows that glass regenerative amplifiers could support pulses as short as 500 fs.

One can also increase the gain (and thus reduce the gain narrowing) by using two heads with identical glass. Although this approach is not as powerful as the previous one, it offers the advantage of higher stability and greater simplicity. Finally, one can preshape the seed spectrum, in order to increase the amplified pulse bandwidth. This approach, which is most successful when the number of roundtrips through the amplifier is limited, can be combined with the approaches outlined above.

Fig. 7 presents a comparison between: (i) our existing regenerative amplifier (single head design, Q98 glass rod), (ii) a regenerative amplifier with two similar heads (each with Q-98 rod) and (iii) a regenerative amplifier with two heads containing different glass types

(with the two central frequencies separated by 6 nm). The simulation assumed that the 1 nJ input pulse had a 40 Å bandwidth with a linear frequency chirp. The pulse is cavity-dumped at a given energy level (4 mJ). Our simulation shows that a regenerative amplifier of type (i) can support a 1.2 ps pulse. This corresponds very closely to our regenerative amplifier operating at low voltage (1200 V). Increasing the gain (by the addition of a second head, or by increasing the voltage of our standard source) allows the regenerative amplifier to support a 760 fs pulse. Finally a regenerative amplifier of type (iii) can support a pulse as short as 500 fs with the appropriate glass combination.

The finite bandwidth of the gain medium introduces some severe gain narrowing as shown above. Other optical elements may also limit our ability to amplify femtosecond pulses. We are particularly concerned by the thin-film polarizer that is always present in our regenerative amplifier. Figure 8 shows the transmission characteristics as function of the wavelength for two types of thin-film polarizers. This data illustrates the bandwidth limiting effect of the polarizers over 50 round-trips. The thin-film polarizer incident angle alignment is also critical as illustrated in Fig. 9.

III. MISCELLANEOUS ISSUES IN CHIRPED PULSE AMPLIFICATION.

Pulse Compression.

We have characterized the Milton-Roy 1200 l/mm ruled gratings presently being used by some of the groups working with CPA sources. Steve Harris' group (Stanford) has reported that this type of gratings has a much higher damage threshold than the American Holographic grating we have been using in the past.

As announced by the manufacturer the efficiency was measured to be higher than 86%. However, in order to reach that level of efficiency the gratings are blazed. The grooves have a triangular shape that modulates the beam spatial profile. In addition, the presence of ghost has been noticed on all the gratings received from Milton-Roy. These ghosts, resulting from a periodic modulation of the groove spacing, impose some additional modulation of the spatial profile. These two effects combined strongly modulate the beam spatial profile. Consequently, it is no longer possible to focus the beam to its theoretical diffraction-limit without some additional form of spatial-filtering. This operation has to be done after compression.

We have also demonstrated a single-grating pulse compression stage. This elegant approach, based on the symmetrical properties between diffraction of order + 1 and -1, is illustrated in Fig. 10. This new design should reduce substantially the cost of the very large compression stage being designed for the LLNL Petawatt laser.

Frequency-doubling of CPA-generated one-picosecond pulses.

We have doubled the one picosecond-pulses generated by our Terawatt source in a KD*P crystal. The doubling efficiency as function of the intensity is plotted in Fig. 11. The resulting pulses have an extremely high temporal peak-to-background intensity contrast ratio.

Intensity contrast ratio measurement and Pulse cleaning.

One of the primary concerns in the generation of ultra-high peak power pulses is to form pulses with an extremely high peak-to-background intensity contrast ratio. Currently, pulse compression ^{24,25,26} is the standard method for generating short pulses in the 1 micron region. As a result of the nonlinearities inherent in this technique, the pulses produced have a relatively poor intensity contrast (10^2 to 10^3). Our current efforts are directed toward developing techniques to accurately measure this contrast and devising methods to improve it. One should note that our latest results shows that modelocked

Ti:Sapphire oscillators should soon provide an alternative solution for the generation of (sub)-picosecond near-infrared pulses.

We have built two instruments to measure the contrast of our pulses. The first instrument is a second-order autocorrelator capable of accurately measuring the contrast of the compressed pulse prior to amplification. This autocorrelator is designed to give a long scan range (>350 ps), and has a high dynamic range (10^6). Using this diagnostic, we have shown that the maximum achievable contrast ratio with our standard fiber/grating compression scheme is limited to $10^3:1$ as shown in Fig. 12. This instrument is unable to differentiate between pre-pulses and post-pulses. Therefore, we have also built a third order correlator²⁷ to measure the contrast of the pulse after amplification. This tool offers a much larger dynamic range, and is capable of determining without ambiguities the exact pulse structure. Using this latest diagnostic, we have measured the pulse contrast after amplification in the regenerative amplifier. A typical pulseshape, as recorded by our third order correlator, is shown in Fig. 13. The use of our two diagnostics allows us to accurately characterize the contrast of our pulses before and after amplification.

Several methods relying on devices whose transmittance is intensity dependent.^{28,29,30} have been proposed to improve the contrast ratio. We are currently pursuing a cleaning technique based on the intensity dependent phase shift present in single mode optical fibers. Our experimental set-up is shown in Fig. 14.

The nonlinear dephasing through the fiber Φ is given by:

$$\Phi = \frac{2\pi}{\lambda} L \frac{1}{3} n_2 (I_x - I_y)$$

(Where L = fiber length, n_2 = nonlinear index of refraction, and I_x and I_y are the intensities along the fiber principal axes).

The transmission as a function of intensity through the second polarizer is given by:

$$I_{out} = I_{in} \sin^2(\Phi/2) \sin^2(2\alpha)$$

By selecting the proper input polarization one should be able to transmit the peak of a pulse while strongly rejecting its wings. Our simulations indicate that this technique should improved the pulse contrast by two orders of magnitude. The intensities that are required for this cleaning effect to occur are sufficient to be associated with a significant amount of self phase modulation. Thus, the output pulse has reduced wings and can also be further compressed. Using this approach, we have generated pulses as short as 200-fs.

V. SUMMARY

(1) We have demonstrated a cw-pumped, actively mode-locked, Ti:sapphire laser capable of producing 1.7-ps pulses, and through external dispersion compensation, 850-fs pulses. Such short pulse durations are not predicted by Kuizenga-Siegman theory and have prompted an investigation into possible new pulse shortening mechanisms. As these results are very preliminary, we believe shorter pulses will be possible as the mechanisms controlling the pulse formation in this laser become better understood. Nevertheless, even in its present form, this laser is suitable for use as a subpicosecond front-end for Terawatt systems. Such systems presently require a fiber compressor to achieve the desired pulse duration of 1 ps which results in limiting the contrast of the system. Our oscillator shows a significant reduction in the pedestal on which the pulse resides when compared to these systems, despite its complicated spectral shape.

(2) We have shown that a combination of gain media could be used in the regenerative amplifier to drastically reduce any gain narrowing effect. Our transmissions measurements of thin-film polarizers show that these optical elements introduce some severe bandwidth limitation.

(3) We have characterized the Milton-Roy ruled gratings presently being used by some of the groups working with CPA sources. The groove profile and the ghosts associated with this type of gratings strongly modulates the beam spatial profile. After compression the beam is no longer diffraction-limited.

(4) We have tabulated the doubling efficiency of our one-picosecond pulse in a 5 mm KD*P crystal. Optimum efficiency was obtained for an intensity of 13 GW/cm².

(5) We have built two autocorrelators with very high dynamic ratio in order to characterize our CPA-pulses before and after amplification. We are pursuing a cleaning scheme that should ultimately improve the intensity contrast ratio of our source by two-orders of magnitude. This scheme provides the added advantage of additional pulse compression. Pulses as short as 200-fs have been generated.

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VII. FIGURE CAPTIONS

- Figure 1. Color-center APM oscillator schematic.
- Figure 2. Color-center oscillator tuning curves.
- Figure 3. Stimulated emission cross-section of Ti:sapphire for s- and p-polarizations. The hatched region corresponds to the wavelength regime in which this work was performed.
- Figure 4. Ti:sapphire oscillator schematic.
- Figure 5. Compressed pulse duration and average output power versus wavelength.
- Figure 6. Spectrum of the pulses injected in the Nd:glass regenerative amplifier (FWHM is 4 nm) and spectrum of the same pulses after amplification in the Nd:glass regenerative amplifier (FWHM is 1.8 nm).
- Figure 7. Bandwidth associated with various regenerative amplifiers.
(i) our existing regenerative amplifier (single head design, Q98 glass rod),
(ii) a regenerative amplifier with two similar heads (each with Q-98 rod),
(iii) a regenerative amplifier with two heads containing different glass types (with the two central frequencies separated by 6 nm).
- Figure 8. Transmittance of two type of thin-film polarizers as function of wavelength (Transmittance calculated for 50 round-trips).
- Figure 9. Burleigh thin-film polarizers transmittance as function of incidence angle (Transmittance calculated for 50 round-trips).
- Figure 10. Single grating pulse compression stage.
- Figure 11. Doubling efficiency for 1-ps CPA-generated pulse.
- Figure 12. Second-order autocorrelation trace of a compressed pulse.
- Figure 13. Third-order autocorrelation trace of a compressed pulse.
- Figure 14. Experimental set-up for fiber-based pulse cleaning stage.

NaCl:OH Color Center Laser Layout

Tuning Curves of KCl and NaCl Color Center Laser

Ti:Sapphire Stimulated Emission Cross Section

Ti:Sapphire Oscillator

Ti:Sapphire Performance

TOP VIEW

Jean-Luc TAPIÉ	Dec. 98
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Second Harmonic Autocorrelation

Experimental Set-up

Fiber: Nonpolarization preserving, birefringent, single - mode, ~ 80 m

MO: Microscope objectives from fiber couplers

$\lambda/2$: Half-wave plates (the tilted one is used as a Soleil-Babinet compensator)

P: Polarizers