Approaching the microjoule frontier with femtosecond laser oscillators

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Abstract. Broadening the ultrashort laser pulse in a Kerr-lens mode-locked laser by net positive round-trip group-delay dispersion has proven to be a powerful concept for scaling the pulse energy directly achievable with a femtosecond laser oscillator without external amplification. Drawing on this concept, we demonstrate here Ti:Sa chirped-pulse oscillators delivering sub-40 fs pulses of 0.5 \(\mu\)J and 50 nJ energy at average power levels of 1 and 2.5 W (repetition rate: 2 and 50 MHz), respectively, which to the best of our knowledge constitute the highest pulse energy and average power achieved with a femtosecond (<100 fs) laser oscillator to date. The 0.5 \(\mu\)J pulses have a peak power in excess of 10 MW and reach a peak intensity >10\(^{15}\) W cm\(^{-2}\) (when focused down to \(\sim 1 \mu\)m\(^{2}\)), both of which represent record values from a laser oscillator. These pulse parameters appear to be limited merely by the pump power available, affording promise of scaling chirped-pulse femtosecond Ti:Sa oscillators to microjoule pulse energies and—by simultaneous spectral broadening—towards peak power levels of several hundred megawatts.

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1. Introduction

Femtosecond laser oscillators delivering pulses with energies at the hundred nanojoule level or beyond are of interest for numerous applications, including frequency conversion of coherent light into the infrared as well as ultraviolet and extreme ultraviolet spectral ranges and precision laser machining of dielectrics on a submicrometre scale. These technologies are important for scientific and industrial applications likewise and have relied on master-oscillator-power-amplifier systems [1]–[4]. Replacement of these complex and expensive femtosecond laser systems with compact and relatively low-cost sources without compromising their efficiency in converting coherent light into spectral ranges where no lasers are available and in machining materials at MHz rates would greatly expedite proliferation of these novel optical technologies. Standard femtosecond laser oscillators deliver pulses with energies of merely a few nanojoules [5], which is insufficient to write waveguide structures in the bulk material (required pulse energies >10 nJ [6, 7]). On the other hand, master-oscillator-power-amplifier systems suffer from low pulse energy stability and correspondingly reduced material processing quality.

The traditional approach of increasing the pulse energy from femtosecond laser oscillators has been cavity dumping [8, 9]. Its complexity and limitations (pulse energies typically below 100 nJ) barred this technique from widespread use. Another, more recent concept is based on a substantial increase of the resonator length [10, 11]. This approach provides room for increasing the pulse energy at constant average power but, in the early demonstrations [10, 11], it was limited in its scalability by instabilities caused by excessive nonlinear effects in the laser medium.

Recently, two different approaches have been successful in overcoming these limitations and opened the door to breaking the 100 nJ barrier in femtosecond pulse generation from laser oscillators. The first concept stabilizes single femtosecond pulse formation by high net negative cavity group-delay dispersion (GDD) and allowed 100 nJ scale pulse generation at the full repetition rate of a mode-locked laser for the first time [12, 13]. An alternative technique draws on a small amount of net positive cavity GDD [14, 15], which tends to broaden the pulse increasingly with increasing pulse energies, thereby permitting scaling to higher pulse energies without the onset of instabilities [12, 16]. Both approaches have their advantages and drawbacks.

The main advantage of the former concept is the generation of chirp-free, near-bandwidth limited femtosecond pulses directly from the laser oscillator. However, the laws of soliton-like pulse formation imply that the pulse duration must be traded off against the pulse energy. In the chirped-pulse mode locking approach, the laser delivers heavily chirped pulses of several picosecond duration, requiring pulse compression external to the cavity but offers potential of scaling the pulse energy without compromising the compressed pulse duration.
In this paper, we report a series of experiments performed with Kerr-lens mode-locked (KLM) Ti:Sa laser oscillators over a wide range of repetition rates in the operational regime of net positive cavity GDD. The record-breaking results suggest that chirped-pulse Kerr-lens mode-locking constitutes a powerful concept for scaling the pulse energy and peak power in mode-locked lasers and affords promise of dramatically pushing the frontiers of femtosecond laser oscillator technology in terms of both average and peak power. In this paper, we summarize our experimental findings, the results of their numerical analysis being presented in a follow-up paper [17].

2. The experimental systems

The schematic layouts of the KLM Ti:Sa laser oscillators used in our experiments are depicted in figure 1. The active medium in the first laser configuration 1a is a 2.6 mm long Ti:Sa crystal exhibiting an absorption of approximately 70% at the pump wavelength of 532 nm (inverse absorption length $\alpha \approx 3.7 \text{ cm}^{-1}$). For configurations 1b and 1c the length of the crystal is 3 mm. A Verdi X (Coherent Inc.) served as a pump source for all three lasers. The resonators are made up of chirped multilayer mirrors exhibiting negative GDD for compensating the positive GDD introduced by the sapphire crystal, the intracavity prisms and air. A pair of Brewster-angled fused silica prisms mounted on translational stages and separated by some 0.5 cm allows continuous fine tuning of the net intracavity GDD at the border between net positive and negative cavity GDD. Optimum output coupling was found to be in the range of 20–30% for the relevant pump power regime of 5–10 W. A double-pass extracavity compressor consisting of LaK21 prisms was used for compressing the pulses delivered by the chirped-pulse KLM lasers.

The basic configuration shown in figure 1(a) constitutes a standard KLM cavity yielding a pulse repetition rate in the range of 50–70 MHz. To scale the pulse energy beyond what is feasible by increasing the pump power, we supplemented this standard cavity by one or two specially designed multi-pass telescope delay lines vastly increasing the cavity length without compromising alignment sensitivity [18, 19]. By changing the number of bounces and distances between the telescope mirrors, we were able to realize a number of different cavity lengths corresponding to repetition rates between 10 and 5 MHz with one telescope and between 3 and 2 MHz with two telescopes. Changing the cavity configurations unavoidably implied changes in the frequency dependence of the net intracavity GDD due to the differences in the GDD curves of different cavity mirrors. Although these changes have been modest, they introduced significant modifications to the mode-locked spectra in the different operating regimes, as revealed by the results presented in the next section.

Our recently reported relevant results (200 nJ, 27 fs) [16] were obtained with the 10 MHz version of the one-telescope system shown in figure 1(b) (ROC = −50 mm). In this work, we improved this previously demonstrated system in several ways to achieve higher pulse energies. We (i) increased the cavity length from ∼15 to ∼75 m by adding a second telescope (figure 1(c)), (ii) expanded the beam waist in the laser crystal by increasing the radii of curvature of the focusing mirrors from −75 to −100 mm and, last but not least, (iii) incorporated a saturable semiconductor Bragg reflector (SBR) into the laser cavity (the SBR was supplied by Femtolasers Produktions GmbH). The laser beam was gently focused (ROC = −300 mm) onto the SBR chip (size: 5 mm × 5 mm × 0.5 mm) glued to a copper-heat sink with a thermoconductive paste. The high reflectivity and low-GDD ranges (deviations of less than ±100 fs$^2$ from zero) of the SBR were limited to a spectral band of approximately 40 nm centred around 800 nm.
Figure 1. Cavity configurations of the realized oscillators. (a) 70 and 50 MHz oscillator. (b) 10, 7 and 5 MHz oscillator. (c) 3 and 2 MHz oscillator. Inset: the beam spots on the delay line mirrors (for the 360 nJ oscillator). All mirror radii are shown in mm. SBR, saturable Bragg reflector; FS, fused silica prisms; and OC, output coupler.
Figure 2. Spectra and autocorrelations of the oscillators in the configuration shown in figure 1(a). Black-line and red-line spectra: 70 MHz repetition rate, no SBR, 30 nJ pulses (7.5 W of pump). In the negative-dispersion regime (red-line spectrum) the pulse duration is 15 fs, see red inset. In the positive-dispersion regime (black-line spectrum) the same mirror combination provides 1.5 ps pulses before the compression and 40 fs after (see left upper inset). Blue-line spectrum and autocorrelation: 50 MHz repetition rate, with SBR, 50 nJ pulses (10 W of pump).

3. Results and discussion

Although almost all system configurations enabled us to start mode-locked operation directly in the regime of positive cavity GDD without the SBR, its presence proved essential for sustaining long-term stability of mode-locking for repetition rates of 5 MHz or lower. From the start-up and steady-state behaviour of the oscillators equipped with a SBR, we may conclude that in the presence of a SBR saturable absorption in the SBR rather than Kerr lensing in the laser crystal introduces the dominant self-amplitude modulation mechanism in both the transient and steady-state stages of mode-locking. This was evidenced by the insensitivity of operation in both stages of the mode-locking process to the adjustment of the distance of the focusing mirrors within the stability range. Whereas start-up critically relied on adjusting this distance to the borders of the stability zones [20] in the absence of the SBR and required careful optimization, its introduction in the laser has made start-up a routine procedure (small mechanical perturbation to one of the end mirrors) and allowed short-term operation even without isolating the laser from its surroundings by a housing. The SBR has not shown any degradation in its performance over an operation period of some 500 h.

The length of the standard cavity containing no telescope (figure 1(a)) was 3 m (repetition rate: 50 MHz) in our experiments. The net intracavity GDD could be readily tuned to negative and positive values by translating one of the intracavity prisms. Figure 2 summarizes results representing routinely reproducible performances in the two (soliton-like versus chirped-pulse) regimes of mode locking. In the regime of negative GDD (soliton-like mode-locking) with the SBR removed from the cavity the laser delivers near-bandwidth limited 15 fs pulses. In the case...
of net positive cavity dispersion (chirped-pulse mode-locking) the laser yields heavily chirped \( \simeq 1.5 \text{ ps} \) pulses, which can be compressed by the extracavity prism pair to a duration of 50 fs, which closely approaches the Fourier limit for the 30 nm broad spectrum. In the regime of negative GDD the increase of output pulse energy with pump power is stopped at a pump power level of around 5.5 W by the onset of instabilities limiting the maximum output pulse energy to \( \simeq 10 \text{ nJ} \) (output coupling: 23%). In sharp contrast, adjustment of the cavity GDD to a sufficiently large positive value (\( \simeq 100 \text{ fs}^2 \)) and use of a 30% output coupler enabled us to increase the pump power up to its maximum value of 10 W without compromising the stability of mode-locked operation. This pump power resulted in an average output power of 2.5 W, implying a pulse energy of 50 nJ. No evidence of saturation of the output average power was observed up to 10 W of pump power.

By adding one or two telescopes to the standard cavity configuration, we were able to increase the cavity length to 15–30 m, implying a range of repetition rates of 10–5 MHz, and to 50–75 m (3–2 MHz), respectively. Upon increasing the cavity length, we also expanded the beam diameter in the laser crystal by using mirrors of radius of curvature of \(-75\) and \(-100 \text{ mm}\), respectively. In the 10–5 MHz range the qualitative behaviour of the systems was similar to that of the 50 MHz setup. With a suitable setting of the positive cavity GDD, we were able to realize stable mode-locking up to the 10 W pump level, resulting in a maximum output pulse energy of 360 nJ at a repetition rate of 3 MHz (output coupling: 30%). In contrast, the maximum pulse energy was limited to 505 nJ by the onset of instabilities at a pump power level of 8 W in the 2 MHz cavity equipped with a 30% output coupler.

We used several diagnostics for observing a possible break-up of the laser pulse into several pulses and redistribution of intracavity energy into a cw background. These include a high-resolution spectrometer, a long-scan range (\( \pm 300 \text{ ps} \)) autocorrelator and a high-dynamic range fast (response time: 500 ps) photodetector. Figure 3 shows typical behaviour close to the border of unstable operation. Pre-pulse grows in time as the pulse energy exceeds the threshold. Note that there is a gap between the main pulse and pre-pulse. Two spikes seen to the right of the main pulse are detector electrical artefacts.

![Figure 3](http://www.njp.org/)
Figure 4. Autocorrelation trace (a), optical spectrum and intracavity dispersion (b) of the 360 nJ pulses. Optical spectrum and dispersion (c) of the 505 nJ pulses. Autocorrelation in both cases looks similar, so is shown only once. Side wings on the autocorrelation indicate remaining uncompensated pulse chirp.

of the stable mode-locking regime in the 2 MHz laser. With a pump power approaching the threshold for the onset of unstable operation, the energy content of a long pre-pulse rapidly increases. Note that the autocorrelation may appear to be stable over a substantial fraction of this regime and is therefore not suited for monitoring this behaviour.

The onset of instabilities in the 2 MHz system can be understood by assessing the peak intensity of the pulse in the laser crystal and related Kerr phase shift. It is found to be comparable
Figure 5. (a) Spectral width as a function of intracavity GDD for 100 nJ 10 MHz oscillator (configuration figure 1(b)) and (b) the generated spectral shapes. Black curve: M-shaped spectrum, red: $\Pi$-shaped, green and blue: parabolic-like spectra. The spectral width increases with decreasing GDD.

to that suffered by the pulse when mode locking tends to become unstable in the negative-dispersion regime. Further expansion of the beam diameter is expected to allow us to overcome this limitation.

Figure 4 summarizes the autocorrelation and spectra for the 360 and 505 nJ pulses (with the former being comparable and hence shown only once). Again the intracavity pulse duration was found to be around 1.5 ps. In spite of its vastly increased cavity length, the 2 MHz oscillator appears to be just as stable as the standard high-rep-rate systems, owing to the imaging property of the telescopic delay line. In a housing, it operates for several hours without power degradation. The 0.5 $\mu$J pulse energy and sub-50 fs pulse duration yield a peak power in excess of 10 MW. We managed to focus this laser beam to a spot diameter of $\approx 1 \mu$m with an aspheric lens (Thorlabs, NA = 0.55), implying a peak intensity of approximately $10^{15}$ W cm$^{-2}$ at focus. To the best of our knowledge, these results constitute record values for a femtosecond oscillator.

Variation in the spectra for the different parameter regimes can be attributed to differences in the frequency dependence and nominal value of the net intracavity dispersion. The former is caused by significant differences in the dispersion curves of our available chirped mirrors. On the other hand, even for a given spectral dependence of GDD a change in its nominal value (by changing the path length through one of the intracavity prisms) has a dramatic effect on both the shape and width of the steady-state mode-locked spectrum, as revealed by figure 5. We observed that the pulse chirp rapidly increases with the intracavity dispersion in a way similar to the experimental results in [14]. The autocorrelation shape is sensitive to the dispersion variations as well. This behaviour is to be expected and has also been supported by numerical simulations [17], but has not yet been observed in lasers. Figure 6 illustrates compressibility of pulses with different spectra: autocorrelations shown in figures 6(a) and (b) indicate some remaining uncompressed chirp, while as shown in figure 6(c) the chirp has been completely compensated (no wings on the autocorrelation).

The optimum pulse parameters which we were able to achieve in the experiments described above are summarized in table 1 and figure 7. The broader spectra and the corresponding shorter pulse durations attained with the 10 and 7 MHz systems may be attributed to the absence of
Figure 6. Compressibility of pulses with different spectra: (a) and (b) autocorrelations indicate some remaining uncompressed chirp caused by the sharp spectrum edges; (c) the chirp has been completely compensated (no wings seen on the autocorrelation). (Data provided by A Assion.)

the SBR in the cavity. The significant differences in the mode-locked bandwidth with and without SBR suggests that the use of broadband SBRs may open the way towards chirped-pulse mode locking over a 100 nm bandwidth and beyond. We should note that increasing pulse energy tends to shift the amount of positive GDD required for stable chirped-pulse...
Table 1. Parameters of the realized oscillators. Spectral width is measured on the base of the spectra.

| Cavity length (m) | Repetition rate (MHz) | Cavity configuration | Spectral width (nm) | Pulse duration (fs) | Pulse energy (nJ) |
|------------------|-----------------------|----------------------|---------------------|---------------------|-------------------|
| 3                | 50                    | Figure 1(a)          | 25                  | 50                  | 50                |
| 15               | 10                    | Figure 1(b)          | 80                  | 30                  | 200               |
| 21               | 7                     | Figure 1(b)          | 80                  | 35                  | 250               |
| 50               | 3                     | Figure 1(c)          | 40                  | 40                  | 360               |
| 75               | 2                     | Figure 1(c)          | 35                  | 45                  | 505               |

Figure 7. The realized Ti : Sa oscillators. For each oscillator the spectral width is shown together with pulse duration and necessary negative GDD of the compressor to achieve chirp-free pulses.

mode-locking towards higher values. This experimental observation has also been backed by numerical simulations [17].

In spite of the differences in the spectral dependence of the cavity GDD in the realized chirped-pulse oscillators and related differences in the mode-locked spectral distributions, figure 7 reveals a clear trend: the scalability of the pulse energy with the cavity round-trip time and pump power ensured that the beam size in the crystal and the positive cavity GDD are properly chosen. The results presented afford promise of breaking the microjoule barrier with a Ti:Sa femtosecond laser oscillator in the near future.

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