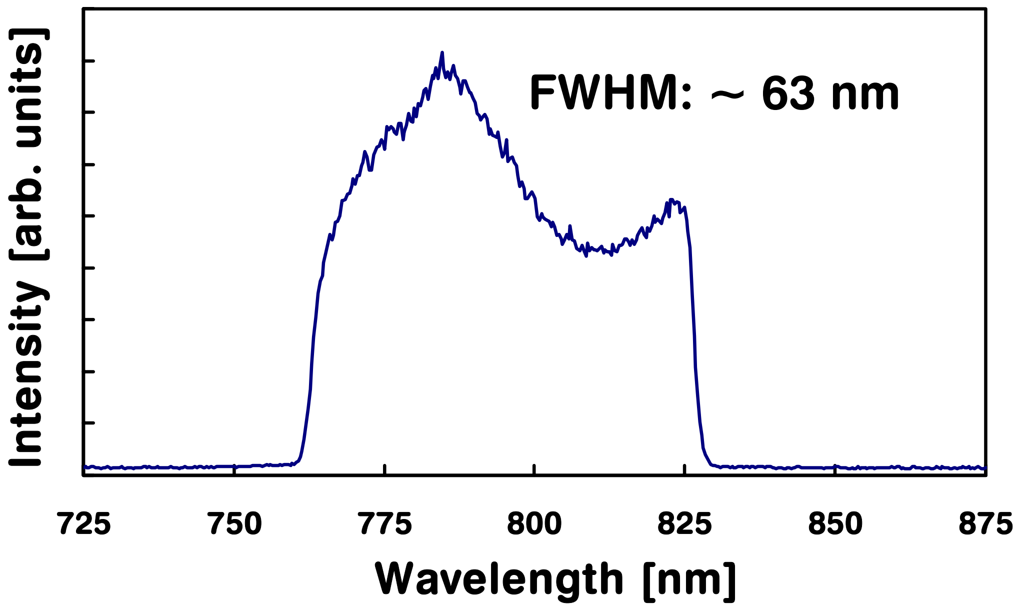
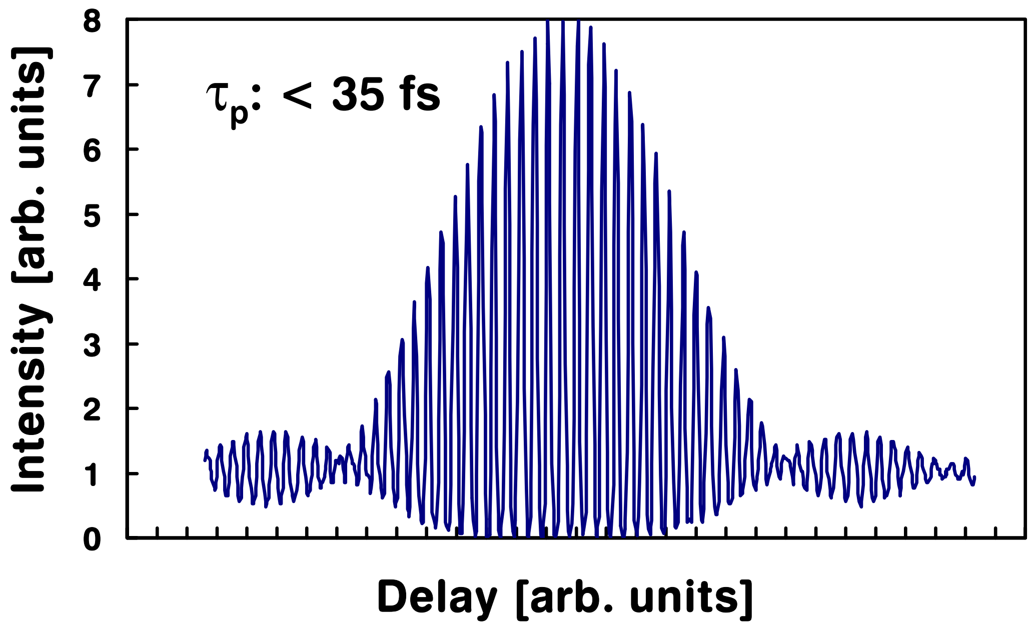
Spectrum of FS Scientific XL



Autocorrelation of FS Scientific XL



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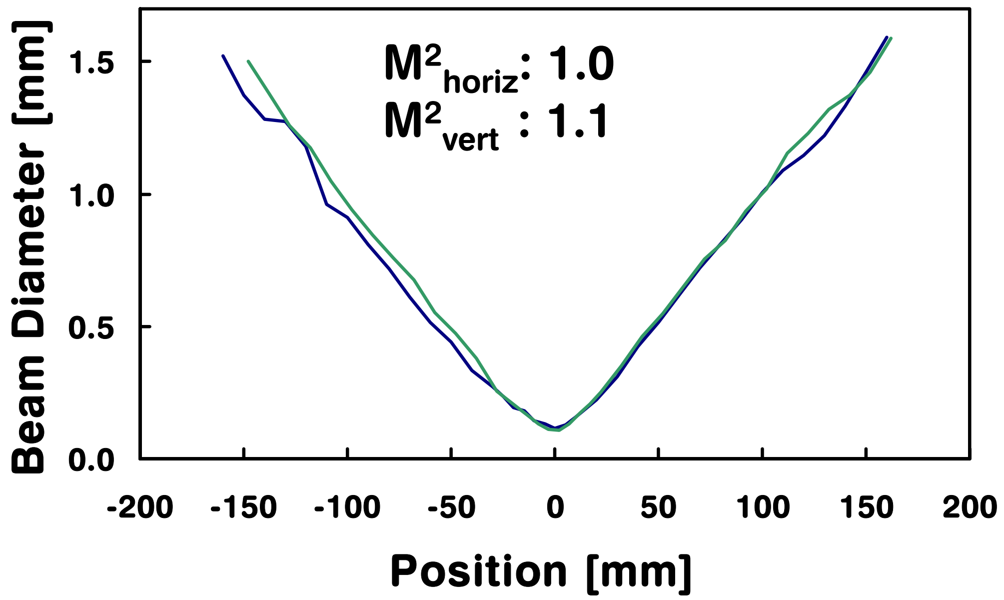
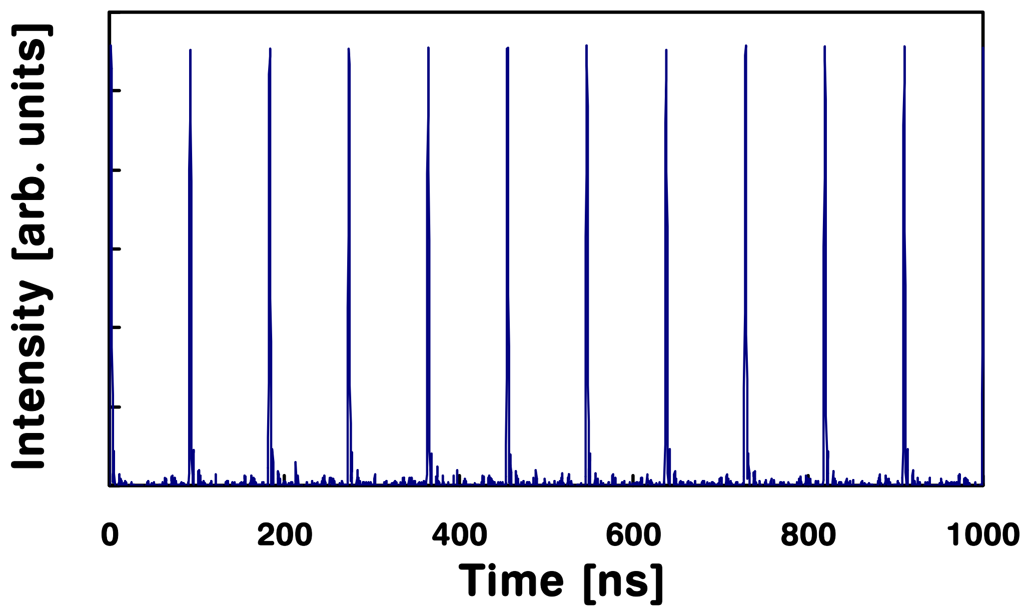
Beam Quality of FS Scientific XL



Pulse to Pulse Stability of FS Scientific XL



Pulse to Pulse Peak Stability: better than 0.38%
(standard deviation measured over 1000 shot)

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Proceedings of SPIE Vol.5340 *Commercial and Biomedical Applications of Ultrafast Lasers IV*
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Generation of sub-30-fs pulses from a scaleable high-energy oscillator

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ABSTRACT

We report on the generation of sub-30fs pulses from a mirror-dispersion-controlled (MDC) Ti:sapphire oscillator, containing a multiple-pass Herriott-cell for increasing the cavity length. Using that scheme, repetition rates down to some few MHz could be achieved. To avoid multiple pulsing instabilities, we operate the laser in a regime of slight positive group-delay dispersion (GDD) over a very broad wavelength range. This results in the formation of strongly chirped light pulses, reducing the otherwise very high peak-intensity inside the laser crystal, which would limit the maximum output energy. We have investigated the spectral phase associated with these pulses with the help of the well known SPIDER-technique, and, based on the results, have constructed an optimized compressor.
When pumped with the full 10 W of a frequency-doubled Nd:YVO_4 laser (Coherent Verdi V10), output energies well above 200 nJ could be obtained. As no signs of instabilities were observed, we believe, that our approach is scaleable to even higher energies if more powerful pump lasers are used.
Thanks to the excellent beam profile, high-resolution micromachining of various materials, including transparent dielectrica could be demonstrated.
Results on sub-micrometer surface modification of transparent materials will be presented.

Keywords: femtosecond, ultrafast, long cavity oscillator, micromachining, ablation.

1. INTRODUCTION

Ultrashort laser pulses with energies exceeding the 100nJ level are very useful for a wide range of applications. From the scientific point of view, there is great interest in the generation of phase-stabilized powerful pulses for nonlinear experiments (generation of high harmonics, photoemission, above-threshold ionization, etc.)[1]. The most important technical applications include 3D binary data storage[2], waveguide writing[3] and nanostructure fabrication in glasses[4].
The generation of very powerful light pulses is usually realized using a low-energy, high repetition rate oscillator, a pulse picker and a subsequent amplifier stage. Therefore such systems are complex, inherently expensive and operate at repetition rates in the few-kHz range or even below. A much more elegant way of producing femtosecond pulses with energies up to hundreds of nJ is the construction of a low repetition rate oscillator.
The pulse energy, out of a mode locked oscillator can be calculated by

$$W_P = \frac{P_{Avg.}}{f_{Rep.}} \tag{1}$$

where $P_{Avg.}$ is the average output power and $f_{Rep.}$ is the repetition frequency.
Therefore, by reducing the repetition rate, which is normally around 100 MHz, much higher output energies can be obtained while the average output power and also the pump power is kept constant. The reduction of the repetition frequency is equivalent with an increase of the resonator length, which can be achieved very efficiently by inserting a multipass cell[5] into the laser cavity. In that way, the resonator can be increased to an effective length on tens of meters, corresponding to a repetition rate of only a few MHz, while keeping the total footprint of the laser small.

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Known limitations of this long-cavity approach are pulse energy instabilities, extra noise at a high peak intracavity intensities and, above all, the onset of multiple pulsing. Two different techniques have already been suggested in order to maintain single-pulse operation even at very high energy levels: to implement a highly negative net-intracavity dispersion[6], resulting in a relatively low spectral bandwidth, or a positive one in a prism-controlled Ti:Sapphire oscillator[7].

The best results in terms of pulse energy have been published recently and constitute the generation of 150 nJ 43 fs pulses at a 6 MHz repetition rate[6]. However, no experiments where these pulses are used have been demonstrated, up to now. As the diagnostic check if all the energy is really concentrated in a single output pulse and no pulse splitting has taken place is very difficult we believe, that only an experimental setup, which depends on the high pulse energy level, can provide absolute evidence in that respect.

Another, yet more complex approach to achieve above-hundred nanojoule fs-pulses at a high repetition rate has been demonstrated recently with a system containing a cavity dumped Ti:Sapphire oscillator plus a subsequent continuous-wave (cw) amplifier[8].

In this paper we present the generation of sub-30-fs pulses at a 200 nJ level directly from a mirror-dispersion-controlled (MDC) Ti:Sapphire oscillator at an 11 MHz repetition rate. The results on surface ablation in transparent media (glasses) with these pulses are also presented.

2. SETUP

Different oscillator schemes have been experimentally tested in order to find the most reliable one capable of generating stable pulses in the several-hundred nanojoule range. All the tested resonators contained no intracavity elements other than the gain medium (a 3mm long Ti:sapphire rod with an absorption of 67% at 532nm), chirped or standard high reflecting dielectric mirrors and a variable amount of fused silica for introducing an additional and tunable amount of positive dispersion. For focusing the beam into the crystal we used different focusing mirrors with a radius-of-curvature (ROC) from 50 mm to 100 mm. As a pump source, a frequency-doubled Nd:YVO_4 laser (Coherent Verdi V10) with an output power of up to 10 W was used. By means of different multipass cell configurations, resonator lengths from 2 m to 21 m, corresponding to a repetition rate from around 7 MHz to 75 MHz were realized.

Full attention was paid, to precisely and fully characterize the temporal structure of the pulse train. In the time range down to some hundreds of picoseconds, a fast photo detector in combination with a fast oscilloscope was used. The pulse-to-pulse stability was also analyzed by that means. For a check on multiple pulses, potentially separated at an even smaller time interval, a modified interferometric autocorrelator with an enhanced scan range of one of its arms was used. The time range from some femtoseconds to hundreds of picoseconds could be covered with that device. To completely characterize the pulses, the well known SPIDER-technique was employed. In addition, the spectral shape of the generated pulses was monitored by an Ocean Optics spectrometer.

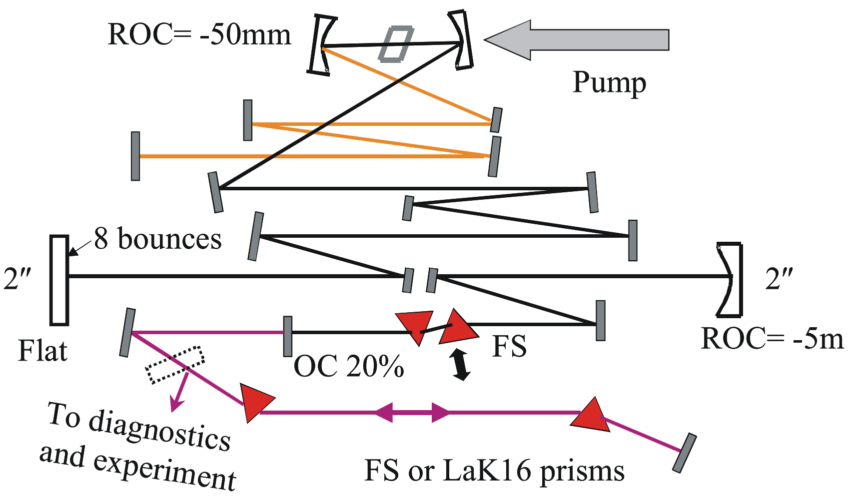

Fig. 1. Experimental Setup of the extended cavity oscillator

All our experiments with extended cavity oscillators operating at a net-negative intracavity dispersion and a high pump power level revealed the formation of additional pulses, separated either 0.8 - 3 ps from the main pulse or some tens of nanoseconds, depending on the intra-crystal intensity and the spectral dispersion distribution. Moreover, after increasing the pump power, we sometimes could also observe a splitting of the single pulse, circulating inside the cavity at moderate pump powers, into one relatively powerful pulse and a large number of small pulses. In total, these satellite pulses can carry a considerable amount of energy, but as each pulse is orders of magnitude smaller than the main pulse, they are very hard to detect. Additionally, the formation of extra noise, easily visible at the interferometric autocorrelation trace was sometimes present in that regime. It should be pointed out, that a picosecond interpulse separation has already been observed experimentally as well as in numerical simulations and can be explained due to a soliton pair equilibrium[9].

Introducing at a net-positive intracavity dispersion resulted in the generation of a much more stable pulse train compared to an oscillator operating at a net-negative dispersion at the same output power level. However, this feature usually comes with the drawback of a reduced spectral bandwidth. But in contrast to low-energy oscillators we found out that in our case, due to the enhanced self-phase modulation (SPM), spectra as broad as almost 100 nm can be generated, if a well designed combination of chirped mirrors and fused silica wedges is used. With that means, the GDD can be kept at a more or less constant, and only slightly positive value, over an extended wavelength range.

The optimized and final oscillator layout is shown in Fig. 1. Two curved mirrors with an ROC of 50 mm, high reflective for the laser wavelength, centered at 800 nm and high transmittive for the pump wavelength of 532 nm are used, to focus the resonator beam into the Ti:sapphire crystal. The total GDD is controlled by various bounces off different chirped dielectric mirrors and two intracavity fused silica wedges. The multipass cell consists of two mirrors with a diameter of 2 inches, one of them a flat mirror whereas the other one a concave type with a ROC of 5 m. There are in total 16 reflections per round trip realized on each of these telescope mirrors which are separated by approximately 73 cm to preserve the q-parameter. This results in an effective resonator length of 13.6 m, tantamount to a repetition frequency of 11 MHz.

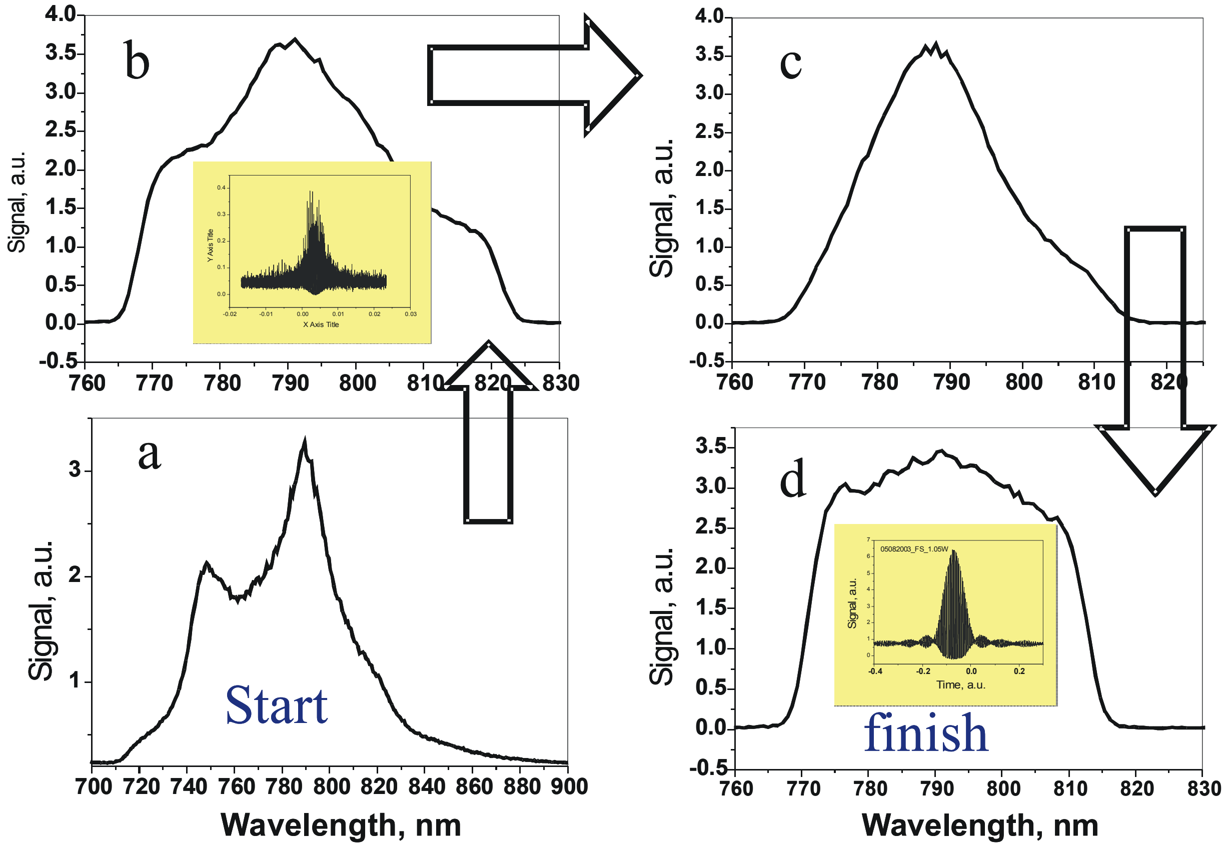

Fig. 2. Evolution of the spectral intensity for an increasing amount of net-GDD inside the cavity.
Inlet: the autocorrelation trace in a "noisy" (4b) and a "stable" mode (4d).

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3. RESULTS

Depending on the amount of the averaged net-intracavity dispersion, which can be fine-tuned by changing the insertion of one of the fused silica wedges, different modes of operation could be observed. Starting at slightly net-negative GDD values (Fig. 2a), the laser emits near-diffraction limited femtosecond pulses with strong pulse-to-pulse fluctuations. Careful analysis of the pulse train shows, that pulse splitting has occurred and part of the pulse energy has been transferred from the main pulse to several satellite pulses. When a larger amount of fused silica is introduced into the resonator and the net-dispersion is therefore shifted towards positive values, the pulses start to become strongly chirped and all the energy is now concentrated into a single high energetic pulse. However, if one tries to compress this pulse train by an extracavity prism compressor, only a very noisy autocorrelation trace can be obtained (Fig. 2b). A further increase of the amount of the (now positive) net-intracavity dispersion, results in a significant change of the spectral shape (Fig. 2c), until a near-rectangular spectral intensity is measured (Fig. 2d). Compressing these pulses by means of the extracavity prism-pair, a clean autocorrelation trace and a very high pulse-to-pulse stability can be obtained now. The right part of Fig. 3 shows the corresponding output pulse train, measured with a fast photo diode. The maximum variations in the pulse energy are well below 3%. As a comparison, on the left, a typical pulse train out of a high energy oscillator, operating at a very low absolute value of net-negative intracavity dispersion is shown. Pulse splitting and instabilities are an unavoidable feature in this regime.

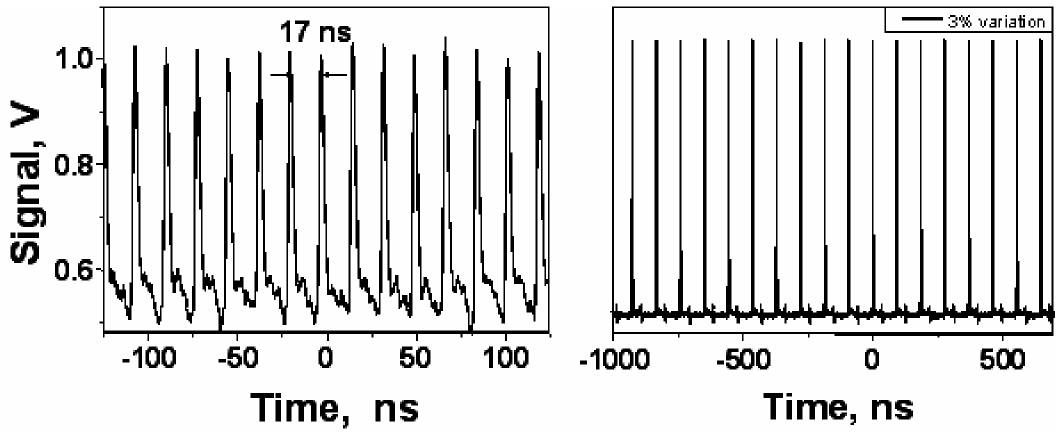

Fig. 3. Pulse train in the slightly negative (left) and slightly positive dispersion regime (right).

The heavily chirped picosecond pulses, emitted by the oscillator when operated at positive values of the net-GDD were, as already mentioned above, compressed by means of an extracavity compressor consisting of a pair of either SF10, fused silica or LaK16 prisms. The use of SF10 as prism material allowed to build a compact (~75 cm apex-apex distance) compressor, but the quality of the compressed pulses was poor, showing a large amount of uncompensated high order phase terms. The best result in terms of pulse quality could be obtained using the fused silica compressor. Unfortunately, a very large prism separation of almost 5 m is necessary in that case, impairing the beam pointing stability of the system. As a compromise, we finally used a 1.4 m long compressor consisting of two LaK16 prisms.

Further optimizing the spectral distribution of the overall intracavity GDD, by using chirped mirrors with different designs, we were able to demonstrate an even broader output spectrum with a FWHM-width of more than 70 nm. However, even with the fused silica prism compressor, the pulses could not be fully compressed. SPIDER measurements revealed, that the spectral wings still contained a non-compensated chirp equivalent to a GDD of around 10^4 fs^2 . We therefore additionally introduced several bounces off special chirped dielectric mirrors with a high third-order dispersion (TOD). As a result, near transform-limited pulses with a pulse duration as short as 27 fs could be obtained. Figure 4 shows a complete characterization of these output pulses, including, besides the autocorrelation trace and the spectral intensity, also the results of the SPIDER-measurement. One has to take into consideration, that for such an unusual spectrum with sharp edges only SPIDER or FROG measurements can provide true information about the pulse shape and duration.

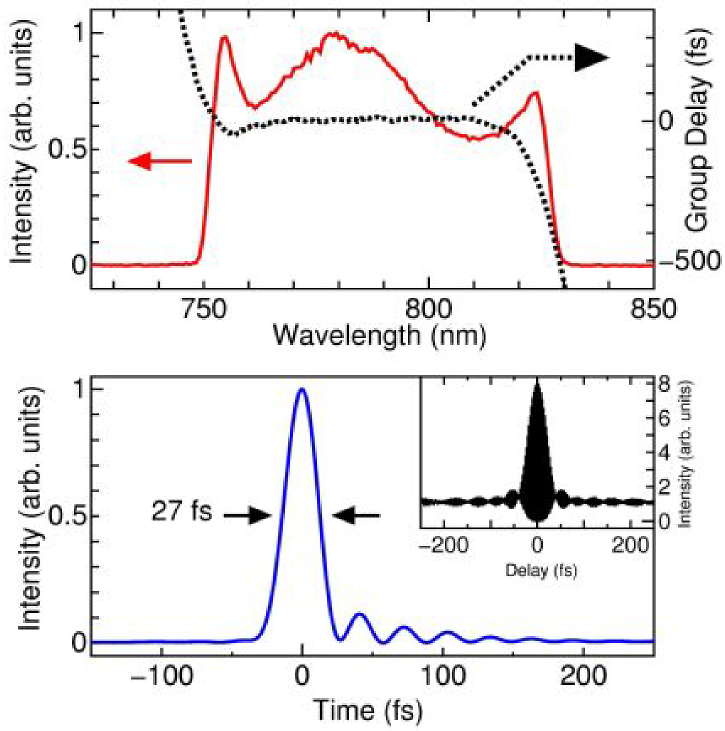

Fig. 4. Spectral intensity and phase, based on a SPIDER measurement (above).
Below: Retrieved pulse shape and measured autocorrelation function (inset).

We found out, that the maximum available pump power (up to 10.1 W) could be applied to the oscillator without any signs of multiple pulsing or continuous-wave (cw) breakthrough. The largest output power of 1.9 W directly out of the oscillator was obtained using an output coupler (OC) with a transmission as high as 29%. This is, to our knowledge, by far the highest average output power ever generated with an extended cavity Ti:Sapphire oscillator. Inserting an output coupler with a still higher transmission resulted in an even higher cw output power, but we were unable to initiate mode-locking. As we used Brewster prisms for compressing the pulses, the transmission losses could be kept below 5%. Taking into account the repetition rate of 11 MHz, this corresponds to a pulse energy of 165 nJ behind the compressor. Because the full available pump power could be used without any indications of limitations or saturation, we believe that our approach is scaleable to even high average powers by using stronger pump lasers or by pumping the gain medium from both sides.

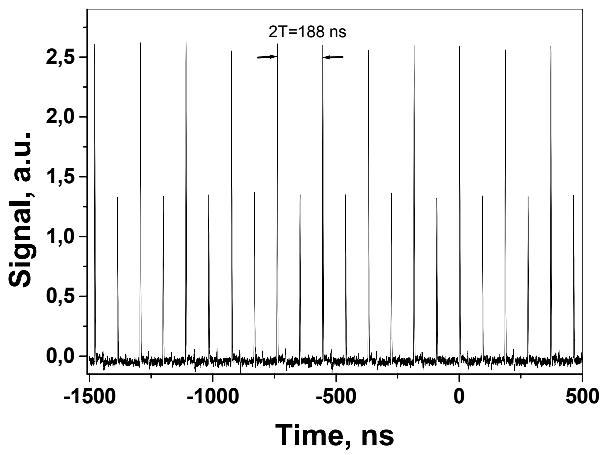

Fig. 5. Regime of period doubling. The power ratio between subsequent pulses can be as high as 3:1.

Depending on the position of the Ti:sapphire crystal relative to the resonator beam and the exact position within the stability range, an interesting operation regime was found, where every second pulse contains a larger amount of energy and a lower one, respectively (see Fig. 5). This regime of so-called period doubling results in a significant increase of the pulse energy of every second pulse. In contrast to observations made in a standard oscillator[10], we did not see any difference in the beam size between the normal operation regime and the period doubling. Unfortunately, this regime depends very critically on the overall cavity alignment and the crystal position. However, once it is established, it remains stable over a long period of time. From the pulse train shown in Fig. 4, a pulse energy of the stronger pulses as high as 220 nJ can be derived. The period doubling regime can be of great interest for nonlinear scientific experiments and material processing applications.

4. MATERIAL PROCESSING

There are a couple of reasons, making a long cavity, high energy Ti:sapphire oscillator the perfect tool for micromachining applications in transparent dielectric materials. First of all it is known, that the damage threshold in such materials becomes lower for shorter pulses[11,12]. Therefore, less energy has to be deposited into the material, minimizing the debris deposition around the fabricated structure and maximizing the quality of the process. Additionally, the damage threshold changes from a stochastic behavior for long pulses to a deterministic one for short pulses[11], allowing to adjust the laser energy in a way, that only the center of the transversal intensity distribution exceeds the ablation threshold. In that way, structures even smaller then the size of the focal spot can be generated. On the other hand, an ultrahigh precision usually comes with the drawback of a low efficiency of the overall machining process, because only a very small volume is ablated per laser shot. As the presented oscillator operates at a repetition rate in the MHz range, compared to amplifier systems, usually operating at a few kHz, much higher ablation rates can be achieved.

To avoid additional (and usually parasitic) surface modifications around the structures which one actually wants to generate, thermal diffusion of heat out of the focal volume must overcome the deposited laser energy. If this is the case, no temperature raise around the focal area takes place, and no cumulative effects are expected. This ensures, that the machining precision after many laser shots does not degrade in comparison to a single pulse damage spot. To estimate the highest repetition rate of the pulses irradiating the material, meeting this demand, one has to solve the Fourier diffusion equation. Carslaw et.al.[13] proposed a one-dimensional solution of the equation which gives a specific diffusion time scale for glasses approximately equal to 1 µs. Therefore if the repetition rate of the laser is higher than the inverse of this time, namely 1MHz, the generation of nanostructures will only be possible, if the sample is moved fast enough, so that each laser pulse irradiates a different spot. Otherwise, cumulative effects will cause the damage site to enlarge. We therefore plan to further reduce the repetition rate of the long cavity laser. One has to keep in mind, however, that a 1MHz repetition rate already corresponds to a resonator length of 150 m, which is hard to realize.

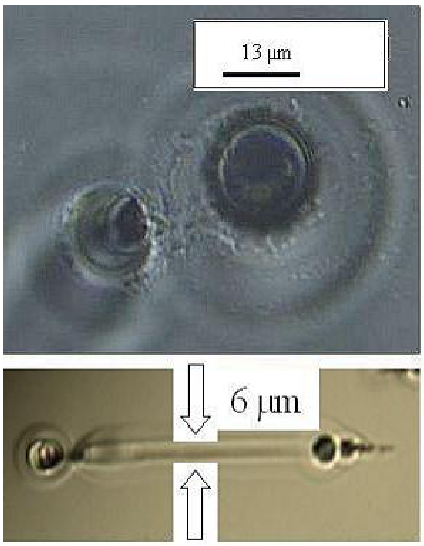

Fig.6. Laser-generated structures on a BK7 substrate: A tapered hole and a groove.

To prove if all the energy is really concentrated in a single output pulse in our laser, and that no pulse splitting has occurred, we performed ablation experiments in glass samples. The pulses behind the compressor were focused onto the surface of a BK7 substrate by means of different optics. Even by using a lens with a numerical aperture (NA) as low as 0.15, surface damage could be achieved. Taking into account the given the damage threshold of BK7 at a 30 fs pulse duration[11] this is a clear evidence, that the pulse energy must be significantly larger than 150 nJ, proving the single pulse operation of the oscillator.

Using an aspheric lens with a NA of 0.4 for focusing the beam onto the same sample, a burst of white-light generation and simultaneous damage was observed (see Fig. 6). The upper pictures shows a conical hole with the input diameter broadened to 13 µm which was obtained after applying a large number of laser shots onto the same spot at the surface of the BK7 substrate. The depth of the hole is about 20 µm. The structure shown at the bottom picture was obtained by moving the sample very fast transversal to the beam. As heat accumulation has a much weaker effect on the ablation process in this case, a grooving with a thickness of around 6 µm was generated.

By carefully adjusting the pulse energy just slightly above the damage threshold and by selecting single output pulses using an extracavity Pockels cell, we tried to generate damage spots with a diameter as small as possible, using the same focusing lens already mentioned above. Because only a standard light-optical microscope was available, an exact analysis of the resulted structure was not possible. However, from the measurement we can estimate, that circular ablation spots with a diameter of around 500 nm were generated on the surface of the BK7 substrate, showing the potential of this laser source for nanomachining applications.

5. CONCLUSIONS

In conclusion, a powerful 220 nJ, 27 fs Ti:sapphire oscillator has been demonstrated at an 11 MHz repetition rate. It provides a robust and stable source of high energy pulses for a variety of applications, most notably micro- or nanomachining of virtual any material. The concept of operating in the regime of positive net-intracavity dispersion seems to be up-scalable, at least no limitations like those in the regime of negative net-dispersion, were pulse splitting has already been observed at moderate pulse energies, have been found. As a direct proof that all the pulse energy is concentrated in a single pulse, ablation experiments on BK7 substrates, using low numerical aperture optics, have been performed. If cumulative effects are neglected, the generation of sub-µm structures becomes possible, due to the short pulse duration.

Our approach does not rely on a saturable Bragg reflector (SBR) to start and maintain mode-locking in contrast to different high energy oscillator shemes[7], and thus allows the generation of broader output spectra, which are at least not limited by the finite bandwidth of the SBR itself.

The combination of an oscillator and a subsequent prism compressor gives one additional flexibility if dispersive elements in the experimental setup (like a microscope objective, a vacuum window, a Pockels cell etc.) have to be pre-compensated. In that way, it easily becomes possible to obtain chirp-free pulses directly on the target without any additional efforts.

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REFERENCES

1. A. Apolonski, A. Poppe, G. Tempea, Ch. Spielmann, Th. Udem, R. Holzwarth, T. W. Haensch, F. Krausz, "Experimental access to the absolute phase of few-cycle light pulses", *Phys.Rev.Lett.*, **85**, 740-743, 2000
2. G. Cheng, Y. Wang, J.D. White, Q. Liu, W. Zhao, G. Chen, "Demonstration of high-density three-dimensional storage in fused silica by femtosecond laser pulses", *J. Appl. Phys.*, **94**, 1304-1307, 2003.
3. K. Minoshima, A.M. Kowalevicz, I. Hartl, E.P. Ippen, J.G. Fujimoto, "Photonic device fabrication in glass by use of nonlinear materials processing with a femtosecond laser oscillator", *Opt. Lett.*, **26**, 1516-1518, 2001
4. R.S. Taylor, C. Hnatovsky, E. Simova, D.M. Rayner, V.R.Bhardwaj, P.B. Corkum, "Femtosecond laser fabrication of nanostructures in silica glass", *Opt. Lett.*, **28**, 1043-1045, 2003.
5. D. Herriott, H. Kogelnik, R. Kompfner, "Off-axis paths in spherical mirror interferometers", *Appl. Opt.*, **3**, 523-526, 1964
6. A.M. Kowalevicz Jr., A.T. Zare, F.X. Kaertner, J.G. Fujimoto, S. Dewald, U. Morgner, V. Scheuer, G. Angelow, "Generation of 150-nJ pulses from a multi-pass cavity Kerr-lens modelocked Ti:Al2O3 oscillator", *Opt. Lett.*, **28**, 1597-1599, 2003.
7. S.H. Cho, F.X. Kaertner, U. Morgner, E.P. Ippen, J.G. Fujimoto, J.E. Cunnighan, W.H. Knox, "Generation of 90-nJ pulse with a 4-MHz repetition rate Kerr-lens mode-locked Ti:Al_2O_3 laser operating with net positive and negative intracavity dispersion", *Opt. Lett.*, **26**, 560-562, 2001.
8. R. Huber, F. Adler, A. Leitensdorfer, M. Beuttner, P. Baum, E. Riedle, "12-fs pulses from a continous-wave-pumped 200-nJ Ti:sapphire amplifier at a variable repetition rate as high as 4 MHz", *Opt. Lett.*, **28**, 2118-2120, 2003
9. M.J. Lederer, B. Luther-Davies, H.H. Tan, C. Jagadish, N.N. Akhmediev, J.M. Soto-Crespo, "Multipulse operation of a Ti:sapphire laser mode locked by an ion-implanted semiconductor saturable absorber mirror", *J. Opt. Soc. B*, **16,** 895-904, 1999.
10. D. Cote, M. van Driel, "Period doubling of a femtosecond Ti:sapphire laser by total mode-locking", *Opt. Lett.*, **23**, 715-717, 1998.
11. M. Lenzner, "Femtosecond laser-induced damage of dielectrics", *Int. J. Mod. Phys. B*, **13**, 1559-1578 (1999).
12. A.P. Joglekar, H. Liu, G.J. Spooner, E. Meyhöfer, G. Mourou, A.J. Hunt, "A study of deterministic character of optical damage by femtosecond laser pulses and applications to nanomachining", *Appl.Phys B*, **77**, 25-30, 2003.
13. H.S. Carlslaw, J.C. Jaeger, *Conduction of Heat in Solids*, p256, Oxford University, London, 1959