Femtosecond Cr:ZnS laser at 2.35 µm mode-locked by carbon nanotubes

Nikolai Tolstik[a], Oleg Okhotnikov[b], Evgeni Sorokin[c], and Irina T. Sorokina[a]
[a] Department of Physics, The Norwegian University of Science and Technology, Høgskoleringen 5, 7024 Trondheim, Norway; [b] Optoelectronics Research Center, Tampere University of Technology, Korkeakoulunkatu 10, Fi-33720 Tampere, Finland; [c] Photonics institute, TU Wien - Vienna University of Technology, Vienna, Austria;

ABSTRACT

We report the solid-state Cr:ZnS laser mode-locked by CNT-based saturable absorber. The absorber was deposited on a protected silver mirror used as a high reflector mirror in a standard 250-MHz cavity with chirped mirror GDD compensation. Laser pulses with duration of 61 fs were obtained at 2.35 μm wavelength. The output power was limited at 950 mW, corresponding to the pulse energy of 3.8 nJ. We have demonstrated the longest-wavelength mid-IR CNT-mode-locked laser with record parameters, advancing the carbon nanotube mode-locking technology well beyond 2 μm into the mid-IR.

Keywords: Mode-locked lasers, Cr:ZnS, Carbon nanotubes;

INTRODUCTION

The mid-infrared (mid-IR) wavelength range between 2 and 5 µm is characterized by the presence of strong fundamental and overtone rovibrational absorption bands of atmospheric constituents, vapours and gases, such as carbon monoxide and dioxide, methane, ammonia, NO_x, etc. The ability of broadband solid-state lasers like e.g. Cr:ZnS or Cr:ZnSe to spectrally cover the wide wavelength range, containing all the above bands simultaneously (for ultrashort pulse systems) is the main advantage of these compact laser sources. Generation of ultimately short pulses in the mid-IR (in the wavelength range above 2 microns) is therefore a very desirable albeit challenging aim.

First experimental realizations of self-starting passively mode locked Cr:ZnSe/Cr:ZnS lasers required Sb-based saturable absorbers (SA) for ultrashort-pulse operation (1, 2). Lately it has been demonstrated that carbon nanotube absorbers (CNTA) inserted in the cavity allows producing mode-locking regime of Tm-doped fiber lasers. A ring cavity thulium fiber laser mode-locked with a single-wall carbon nanotube absorber used in transmission produces 1.32 ps pulses at 1.93 μm wavelength (3), the same idea was exploited in linear cavity all-fiber laser (4), as well as later also using the graphene saturable absorber (5, 6). The picosecond scale pulse sosystems benefit from the compact all-fiber design and do not require stabilization and adjustment. On the other hand, femtosecond scale pulse solutions call for implementation of particularly broad gain media, such as e.g. Cr:ZnSe or Cr:ZnS. These media operate at room-temperature and have the largest relative bandwidth of ~45% of the central wavelength of the laser, can provide very high power levels (up to tens of Watt level), ultrashort pulse duration of only several optical cycles retaining the good beam quality and narrow spectral linewidth at the wide tuning range ~ 1800 cm^{-1}-. Potentially, these lasers are capable of generating the single optical cycle pulses and are perfect media for mode-locking using CNTA or grapheme saturable absorbers. However, unlike graphene, the absorption of the CNTA strongly decreases towards longer wavelengths and it has not been yet proven, if it can be used beyond the 2 μm wavelength.

Recently we realized the first graphene mode-locked Cr:ZnS laser (7), generating the shortest to-date pulses in the mid-IR amounting to 41 fs at 250 mW of output power with a nearly 200-nm (FWHM) spectral bandwidth directly from the oscillator. Such pulse duration corresponds to only 5.1 optical cycles at 2.4 μm and is equal to 14-fs pulses on the emission wavelength scale of Ti:sapphire laser. These are the shortest pulses generated in the mid-IR, as well as the shortest pulses obtained with the graphene saturable absorber to-date.

Solid State Lasers XXIII: Technology and Devices, edited by W. Andrew Clarkson, Ramesh K. Shori,
Proc. of SPIE Vol. 8959, 89591A · © 2014 SPIE · CCC code: 0277-786X/14/$18 · doi: 10.1117/12.2040757

In this paper we present the first SWCNT-mode-locked Cr:ZnS laser and compare it with the graphene mirror mode-locked Cr:ZnS laser. Carbon nanotubes were implemented on the surface of the protected Ag-mirror to form the CNT-based saturable absorber mirror. Stable CW mode-locking was demonstrated at the wavelength of 2.35 μm, thus representing the longest wavelength of operation for any CNT-mode-locked laser to date. The laser generated pulses with minimum pulse duration of 61 fs and maximal spectral bandwidth of 85 nm at output powers up to 950 mW.

EXPERIMENTAL SETUP

The experimental setup is shown in Fig. 1. The cavity was arranged according to the classic astigmatically-compensated design for Brewsters-oriented active elements. As an active element 2.5-mm PVT-grown Cr:ZnS single crystal was used. The concentration of Cr^{2+} ions in the active element was estimated as $6{\cdot}10^{18}$ cm^{-3}. No active cooling was applied to the crystal. As Cr:ZnS exhibits quite high nonlinearity (8), the pulse energy of the femtosecond Cr:ZnS laser is usually limited by the nonlinearity of the active element (9-12). In order to extract maximum average output power from the laser, and at the same time keep the pulse energy under the nonlinearity limit, the cavity length was kept around 120 cm, which corresponds to the pulse repetition frequency around 250 MHz. The laser was pumped by the CW Er-fiber laser providing up to 5 W of polarized collimated output at 1.61 μm. The pump beam was focused onto the crystal by an AR-coated focusing lens (FL) with a focal length of 40 mm. The cavity mode inside the active element was formed by two 50-mm HR mirrors to the spot of about 55 μm diameter. The cavity mode on the saturable absorber was additionally focused by 100-mm-radius mirror to a spot diameter around 130 μm. Output couplers with transmittance in the range 1.8÷18% were tested. The compensation of the group-delay dispersion (GDD) was achieved by two reflections from specially designed dispersive mirror (chirped mirror).

Single-wall carbon nanotubes were synthesized by thermal decomposition of ferrocene vapor in a carbon monoxide atmosphere. A saturable absorber was prepared by a room-temperature dry-transfer process. A piece of nitrocellulose membrane filter with a SWCNT-film was placed on a inch-size protected Ag-mirror with the SWCNT-film upside down. Then, the mirror and the filter were pressed together. After the pressing the membrane filter was peeled off and SWCNT-film was strongly adhered to the mirror surface. The described preparation method is simple and easy scalable. The same saturable absorber was already demonstrated as a mode-locker in Yb-, Er-, and Tm:Ho-doped fiber lasers at wavelengths of 1.05 μm, 1.56 μm and 1.99 μm, respectively (13).

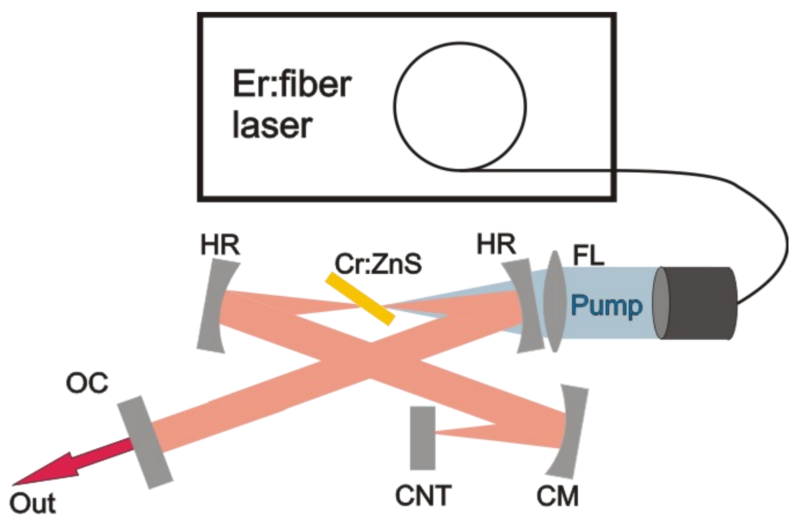

Figure 1. Schematic of the femtosecond Cr:ZnS CNT-mode-locked laser. C – fiber collimator, FL-focusing lens, HR – high reflector mirror, AE – Cr:ZnS active element, CM – chirped mirror, CNT – carbon-nanotubes-based saturable absorber, OC – output coupler.

All measurements were performed in the open air with 30% relative humidity. The spectral distribution of the laser emission was analyzed by a Perkin-Elmer FTIR spectrometer with a spectral resolution of 1 cm^{-1}. For the measurements of the pulse duration a custom-made interferometric autocorrelator was built with a detection system based on a two-photon absorption in an amplified Ge photodetector. The pulse repetition rate was controlled by the picosecond GaAs photodetector working in two-photon absorption regime.

EXPERIMENTAL RESULTS

Stable mode-locking was obtained with all three output couplers. Maximal average output power reached 102 mW, 230 mW and 955 mW for the output couplers with transmission of 1.8%, 5.5%, and 18%, respectively. The input-output characteristics are plotted on Fig. 2. The output power and pulse energy for all three output couplers were, as predicted, limited by the pulse breakup due to excessive nonlinearity of the Cr:ZnS active element The mode-locking was generally not self-started and should be initiated by gentle touching of the saturable absorber holder, though several times we were able to observe the self-starting. The laser was operated near the center of the first stability region, so it is clear that Kerr-lens mechanism was not responsible for supporting of the mode-locking regime.

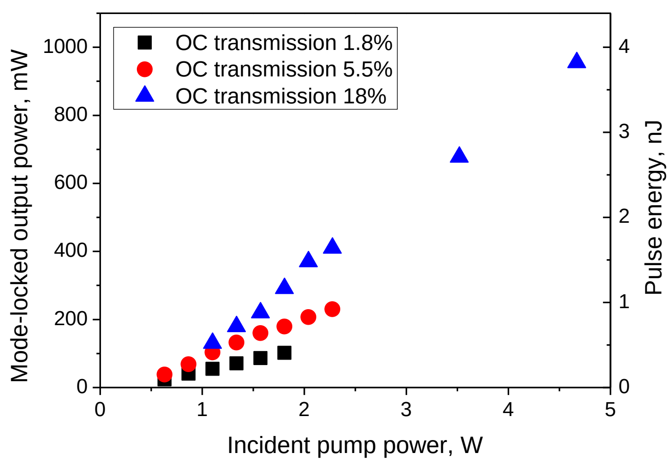

Figure 2. Input-output characteristics of the CNT-mode-locked Cr:ZnS laser with output coupler transmission of 1.8% (black squares), 5.5% (red circles), and 18% (blue triangles).

The pulse duration and spectral bandwidth varied depending on the output power. The evolution of the laser spectrum and the dependence of the pulse duration on the output power for 1.8% output coupler are shown on Fig. 3. The pulse duration, being roughly 450 fs near the mode-locking threshold, reaches the value of 61 fs at the maximum output power of 102 mW. The spectral bandwidth increased from 13 nm near the threshold to 85 nm as a maximum. The similar behavior of the laser characteristics was observed for other output couplers.

The interferometric autocorrelation trace of the shortest pulses having 61 fs duration is plotted on Fig. 4. The pulse duration is equal to 7.5 optical cycles, and corresponds to 20-fs pulses on the wavelength of the Ti:sapphire laser. The laser spectrum and dispersion curve are also shown on the figure. The dispersion compensation by chirped mirror resulted in about -400 fs^2 GDD per cavity roundtrip. The small dispersion-like features on the long-wavelength side of the spectrum are the intracavity traces of the atmospheric absorption (water vapour) (14). The spectrum of the atmospheric absorption is plotted on the same graph for comparison.

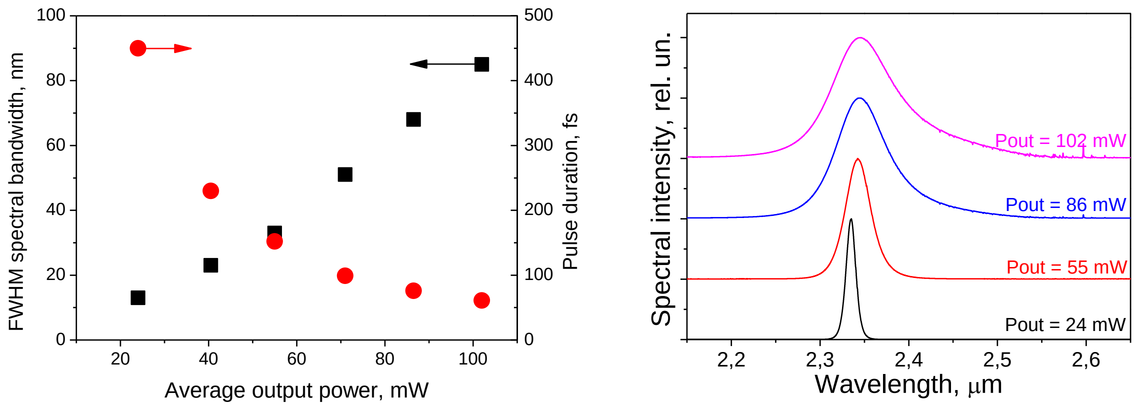

Figure 3. (a) The dependence of the laser emission spectral bandwidth (black squares) and pulse duration (red circles) on the average output power for CNT mode-locked Cr:ZnS laser with 1.8% output coupler; (b) The evolution of the laser spectra on the average output power.

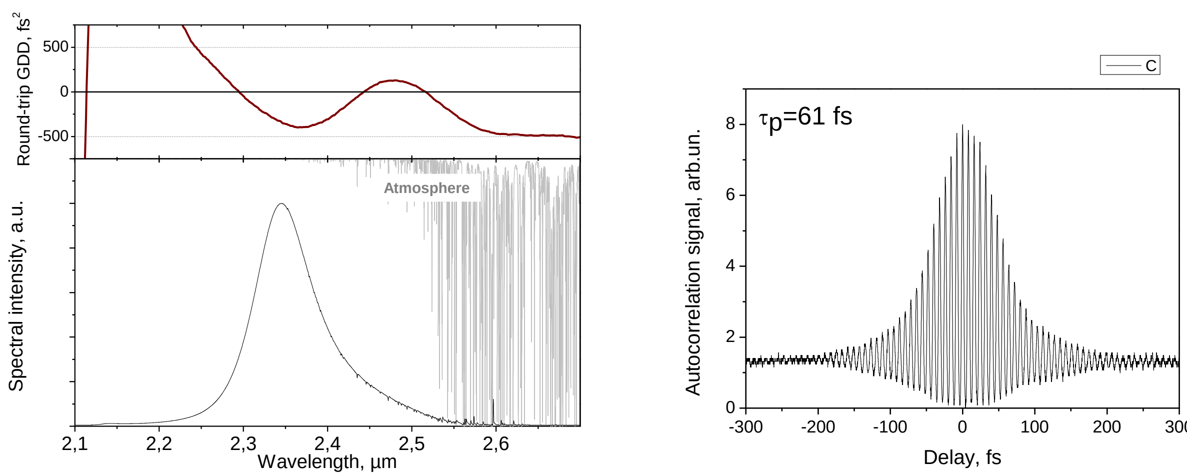

Figure 4. (a) The laser spectrum and round-trip cavity dispersion of CNT-mode-locked Cr:ZnS laser, as well as the atmospheric absorption; (b) The interferometric autocorrelation trace of the shortest pulses with pulse duration of 61 fs.

The unsaturated loss of the CNT-based saturable absorber was measured by a low-power reflectivity measurement to be around 4% with some variations from point to point. The saturated loss was derived from the Caird analysis of the cavity loss (15) in the mode-locked regime to be 5.3 % (Fig. 5). The saturation fluence of the CNT-based absorber mirror was estimated from the mode-locking threshold. Depending on the output coupling rate, the Cr:ZnS laser dropped out of mode-locking at circulating pulse energy not lower than 2.2 nJ. Considering the calculated mode-field diameter of 130 μm on the absorber and coefficient of 2 for Gaussian beam peak power, we estimate the saturation fluence of CNT-based saturable absorber to be around 40 $\mu J/cm^2$.

Another carbon-based saturable absorber for mode-locked lasers is graphene. Graphene mode-locked Cr:ZnS laser was realized recently (16, 17). The output characteristics of both graphene-mode-locked and CNT-mode-locked laser are quite similar with a bit shorter pulses and higher losses for graphene. The pulse duration of the CNT-mode-locked laser could be decreased subject to the implementation of the fine dispersion tuning mechanism, for example YAG wedges (17).

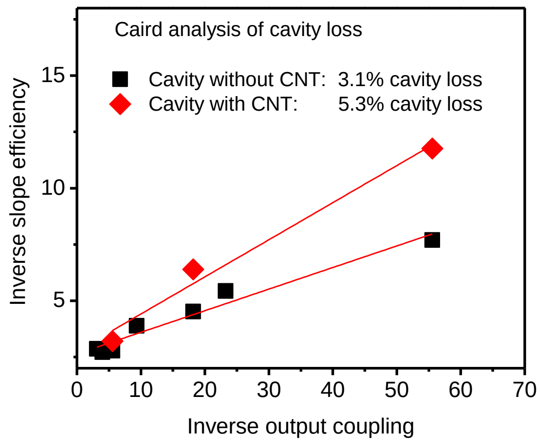

Figure 5. Caird analysis of the cavity loss of Cr:ZnS laser with and without CNT-based saturable absorber..

CONCLUSION

We report the first SWCNT-mode-locked Cr:ZnS laser, confirming the possibility to employ CNT saturable absorbers for mode-locking at wavelengths far beyond 2 μm. We reach pulse duration of 61 fs with a spectral bandwidth of 85 nm at the wavelength of 2.35 μm. The average output power of 950 mW corresponds to the pulse energy of 3.8 nJ. We believe that we report at the same time the longest wavelength of operation, shortest pulses, broadest spectral bandwidth and highest average power generated from any carbon-nanotubes mode-locked laser. Further shortening of the pulse duration and scaling of the pulse energy is feasible, similar to the recently demonstrated graphene-mode-locked Cr:ZnS laser.

ACKNOWLEDGMENTS

This work was supported by the Austrian Science Fund (FWF project P24916) and the Norwegian Research Council (NFR) projects FRITEK/191614, MARTEC-MLR, Nano 2021 project N219686.

REFERENCES

[1] I. T. Sorokina, E. Sorokin, T. J. Carrig and K. I. Schaffers, "A SESAM Passively Mode-Locked Cr:ZnS Laser," in *Advanced Solid-State Photonics*, p. TuA4, Optical Society of America, Vancouver (2006).

[2] I. T. Sorokina, E. Sorokin and T. Carrig, "Femtosecond Pulse Generation from a SESAM Mode-Locked Cr:ZnSe Laser," in *Conference on Lasers and Electro-Optics/Quantum Electronics and Laser Science Conference - CLEO/QELS*, p. CMQ2, Optical Society of America, Long Beach (2006).

[3] S. Kivistö, T. Hakulinen, M. Guina and O. G. Okhotnikov, "Tunable Raman Soliton Source Using Mode-Locked Tm-Ho Fiber Laser," *Photonics Technology Letters, IEEE* 19(12), 934-936 (2007)

[4] M. A. Solodyankin, E. D. Obraztsova, A. S. Lobach, A. I. Chernov, A. V. Tausenev, V. I. Konov and E. M. Dianov, "Mode-locked 1.93 μm thulium fiber laser with a carbon nanotube absorber," *Opt. Lett.* 33(12), 1336-1338 (2008)

[5] K. Kieu and F. W. Wise, "Soliton Thulium-Doped Fiber Laser With Carbon Nanotube Saturable Absorber," *Photonics Technology Letters, IEEE* 21(3), 128-130 (2009)
[6] M. Zhang, E. J. R. Kelleher, F. Torrisi, Z. Sun, T. Hasan, D. Popa, F. Wang, A. C. Ferrari, S. V. Popov and J. R. Taylor, "Tm-doped fiber laser mode-locked by graphene-polymer composite," *Opt. Express* 20(22), 25077-25084 (2012)
[7] N. Tolstik, I. T. Sorokina and E. Sorokin, "Graphene mode-locked Cr:ZnS laser with 42 fs pulse duration," *Opt. Express* (2014)
[8] I. T. Sorokina, "Cr^{2+}-doped II–VI materials for lasers and nonlinear optics," *Optical Materials* 26(4), 395-412 (2004)
[9] N. Tolstik, E. Sorokin and I. T. Sorokina, "Kerr-lens mode-locked Cr:ZnS laser," *Opt. Lett.* 38(3), 299-301 (2013)
[10] E. Sorokin, N. Tolstik and I. T. Sorokina, "1 Watt femtosecond mid-IR Cr:ZnS laser," in *Photonics West*, pp. 8599-8543, San Francisco (2013).
[11] N. Tolstik, I. T. Sorokina and E. Sorokin, "Watt-level Kerr-lens mode-locked Cr:ZnS laser at 2.4 µm," in *CLEO: Science and Innovations*, p. CTh1H.2, Optical Society of America, San Jose (2013).
[12] N. Tolstik, E. Sorokin and I. T. Sorokina, "High-energy Mid-infrared Cr:ZnS Chirped-pulse Oscillator," in *CLEO/Europe*, pp. CF/IE-4.4, Munich (2013).
[13] S. Kivistö, T. Hakulinen, A. Kaskela, B. Aitchison, D. P. Brown, A. G. Nasibulin, E. I. Kauppinen, A. Härkönen and O. G. Okhotnikov, "Carbon nanotube films for ultrafast broadband technology," *Opt. Expr.* 17(4), 2358-2363 (2009)
[14] V. L. Kalashnikov and E. Sorokin, "Soliton absorption spectroscopy," *Phys. Rev. A* 81(3), 033840 (2010)
[15] J. A. Caird, A. J. Ramponi and P. R. Staver, "Quantum efficiency and excited-state relaxation dynamics in neodymium-doped phosphate laser glasses," *J. Opt. Soc. Am. B* 8(7), 1391-1403 (1991)
[16] N. Tolstik, I. T. Sorokina and E. Sorokin, "Graphene Mode-locked Cr:ZnS Chirped-pulse Oscillator," in *Advanced Solid-State Lasers Congress* M. Ebrahim-Zadeh and I. Sorokina, Eds., p. MW1C.2, Optical Society of America, Paris (2013).
[17] N. Tolstik, I. T. Sorokina, A. Pospischil and E. Sorokin, "Graphene mode-locked Cr:ZnS laser with 44 fs pulse duration," in *Advanced Solid-State Lasers Congress* M. Ebrahim-Zadeh and I. Sorokina, Eds., p. MW1C.1, Optical Society of America, Paris (2013).