

90-fs diode-pumped Yb:CLNGG laser mode-locked using single-walled carbon nanotube saturable absorber

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Abstract: A diode-pumped Yb:CLNGG laser is mode-locked with a single-walled carbon nanotube saturable absorber (SWCNT-SA) for the first time. Pulse durations as short as 90 fs are obtained at ~1049 nm with 0.4% output coupler, the shortest pulses to our knowledge for a diode-pumped 1- μ m laser applying SWCNTs as saturable absorber. Using 3% output coupler, the maximum average output power reached 90 mW at a repetition frequency of 83 MHz.

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References and links

1. U. Keller, "Recent developments in compact ultrafast lasers," *Nature* **424**(6950), 831–838 (2003).
2. U. Keller, "Ultrafast solid-state laser oscillators: a success story for the last 20 years with no end in sight," *Appl. Phys. B* **100**(1), 15–28 (2010).
3. H. H. Tan, C. Jagadish, M. J. Lederer, B. Luther-Davies, J. Zou, D. J. H. Cockayne, M. Haiml, U. Siegner, and U. Keller, "Role of implantation-induced defects on the response time of semiconductor saturable absorbers," *Appl. Phys. Lett.* **75**(10), 1437–1439 (1999).
4. T. R. Schibli, K. Minoshima, H. Kataura, E. Itoga, N. Minami, S. Kazaoi, K. Miyashita, M. Tokumoto, and Y. Sakakibara, "Ultrashort pulse-generation by saturable absorber mirrors based on polymer-embedded carbon nanotubes," *Opt. Express* **13**(20), 8025–8031 (2005).
5. W. B. Cho, J. H. Yim, S. Y. Choi, S. Lee, A. Schmidt, G. Steinmeyer, U. Griebner, V. Petrov, D. I. Yeom, K. Kim, and F. Rotermund, "Boosting the nonlinear optical response of carbon nanotube saturable absorbers for broadband mode-locking of bulk lasers," *Adv. Funct. Mater.* **20**(12), 1937–1943 (2010).
6. A. Agnesi, F. Pirzio, L. Tartara, E. Ugolotti, H. Zhang, J. Wang, H. Yu, and V. Petrov, "378-fs pulse generation with the Nd³⁺:SrLaGa₃O₇ (Nd:SLG) disordered crystal," *Laser Phys. Lett.* **10**(10), 105815 (2013).
7. A. Lupei, V. Lupei, L. Gheorghie, L. Rogobete, E. Osiaic, and A. Petraru, "The nature of nonequivalent Nd³⁺ centers in CNGG and CLNGG," *Opt. Mater.* **16**(3), 403–411 (2001).
8. Yu. K. Voronko, A. A. Sobol, A. Ya. Karasik, N. A. Eskov, P. A. Rabochkina, and S. N. Ushakov, "Calcium niobium gallium and calcium lithium niobium gallium garnets doped with rare earth ions - effective laser media," *Opt. Mater.* **20**(3), 197–209 (2002).
9. G. Q. Xie, D. Y. Tang, W. D. Tan, H. Luo, H. J. Zhang, H. H. Yu, and J. Y. Wang, "Subpicosecond pulse generation from a Nd:CLNGG disordered crystal laser," *Opt. Lett.* **34**(1), 103–105 (2009).
10. J. Ma, G. Q. Xie, W. L. Gao, P. Yuan, L. J. Qian, H. H. Yu, H. J. Zhang, and J. Y. Wang, "Diode-pumped mode-locked femtosecond Tm:CLNGG disordered crystal laser," *Opt. Lett.* **37**(8), 1376–1378 (2012).
11. J. H. Liu, Y. Wan, Z. Zhou, X. Tian, W. Han, and H. Zhang, "Comparative study on the laser performance of two Yb-doped disordered garnet crystals: Yb:CNGG and Yb:CLNGG," *Appl. Phys. B* **109**(2), 183–188 (2012).
12. A. Schmidt, U. Griebner, H. Zhang, J. Wang, M. Jiang, J. Liu, and V. Petrov, "Passive mode-locking of the Yb:CNGG laser," *Opt. Commun.* **283**(4), 567–569 (2010).
13. A. Schmidt, S. Rivier, W. B. Cho, J. H. Yim, S. Y. Choi, S. Lee, F. Rotermund, D. Rytz, G. Steinmeyer, V. Petrov, and U. Griebner, "Sub-100 fs single-walled carbon nanotube saturable absorber mode-locked Yb-laser operation near 1 microm," *Opt. Express* **17**(22), 20109–20116 (2009).

14. A. Schmidt, S. Rivier, G. Steinmeyer, J. H. Yim, W. B. Cho, S. Lee, F. Rotermund, M. C. Pujol, X. Mateos, M. Aguiló, F. Díaz, V. Petrov, and U. Griebner, "Passive mode locking of Yb:KLuW using a single-walled carbon nanotube saturable absorber," *Opt. Lett.* **33**(7), 729–731 (2008).
15. A. Agnesi, A. Greborio, F. Pirzio, G. Reali, S. Choi, F. Rotermund, U. Griebner, and V. Petrov, "99 fs Nd:glass laser mode-locked with carbon nanotube saturable absorber mirror," *Appl. Phys. Express* **3**(11), 112702 (2010).
16. J. H. Yim, W. B. Cho, S. Lee, Y. H. Ahn, K. Kim, H. Lim, G. Steinmeyer, V. Petrov, U. Griebner, and F. Rotermund, "Fabrication and characterization of ultrafast carbon nanotube saturable absorbers for solid-state laser mode locking near 1 μm ," *Appl. Phys. Lett.* **93**(16), 161106 (2008).
17. V. Lupei, A. Lupei, C. Gheorghe, L. Gheorghe, A. Achim, and A. Ikesue, "Crystal field disorder effects in the optical spectra of Nd³⁺ and Yb³⁺-doped calcium niobium gallium garnets laser crystals and ceramics," *J. Appl. Phys.* **112**(6), 063110 (2012).
18. H. Zhang, J. Liu, J. Wang, J. Fan, X. Tao, X. Mateos, V. Petrov, and M. Jiang, "Spectroscopic properties and continuous-wave laser operation of a new disordered crystal: Yb-doped CNGG," *Opt. Express* **15**(15), 9464–9469 (2007).
19. C. Fiebig, G. Blume, C. Kaspari, D. Feise, J. Fricke, M. Matalla, W. John, H. Wenzel, K. Paschke, and G. Erbert, "12 W high-brightness single-frequency DBR tapered diode laser," *Electron. Lett.* **44**(21), 1253–1255 (2008).
20. C. Höninger, R. Paschotta, F. Morier-Genaud, M. Moser, and U. Keller, "Q-switching stability limits of continuous-wave passive mode locking," *J. Opt. Soc. Am. B* **16**(1), 46–56 (1999).

1. Introduction

Semiconductor saturable absorber mirrors (SESAMs) have been widely used as saturable absorbers (SAs) for mode-locking solid-state lasers [1,2]. However, while SESAMs exhibit a spectrally narrowband nonlinearity, require sophisticated manufacturing processes and need to undergo post-processing to warrant ultrafast response times [3], single-walled carbon nanotube saturable absorbers (SWCNT-SAs) are characterized by broadband absorption and short recovery time, and can be fabricated by relatively simple methods. Since the first demonstration of steady-state mode-locking with Nd and Er/Yb:glass lasers [4], there have been many reports on passive mode-locking of bulk crystal lasers using SWCNT-SAs, covering the spectral range from ~ 0.8 up to ~ 2.0 μm [5].

Disordered crystals attract more and more attention as hosts for rare-earth ions in mode-locked lasers, because inhomogeneous broadening provides broader spectral emission than common crystals while compared to the even more broadband laser glasses, they exhibit higher thermal conductivity. Moreover, absorption lines in disordered hosts are also broader compared to ordered crystalline matrices, contributing to less demanding stability requirements for the pump diode wavelength. Mostly Nd³⁺-doping has been studied with such disordered crystalline hosts and 378-fs pulses were recently generated with Nd³⁺:SrLaGa₃O₇ using SESAM [6]. Calcium lithium niobium gallium garnet (CLNGG) is a typical disordered garnet crystal in which Nb⁵⁺- and Ga³⁺-ions randomly occupy the same lattice sites. This results in inhomogeneous broadening of the spectral lines of trivalent rare earth dopants. Such dopants substitute Ca²⁺-ions in the lattice and charge compensation imposes specific optimization of the host composition [7]. In order to reduce the concentration of cationic vacancies in calcium niobium gallium garnet (CNGG), which is hard to avoid in the growth process, Li-ions are added. The thermal conductivity of CNGG and CLNGG is the same, 3.5 W/mK, at room temperature [8].

SESAM mode-locking of Nd³⁺:CLNGG produced 900 fs pulse durations at 1061 nm [9], much shorter compared to other Nd-lasers based on ordered crystalline hosts. Recently, 479 fs pulses were obtained with Tm³⁺:CLNGG, again mode-locked by SESAM [10]. We are not aware of mode-locking reports on Yb³⁺:CLNGG. Yb³⁺-doped crystals exhibit strongest electron-phonon coupling and broadest spectral lines among the rare-earth dopants and this spectral broadening mechanism often overwhelms inhomogeneous broadening in disordered crystals. Recently, Yb:CLNGG was compared to Yb:CNNG under diode pumping and showed superior slope efficiency (74% against 63%) and output power (7.5 W against 6.3 W) under identical conditions [11]. Moreover, the larger cross sections and shorter lifetime of Yb:CLNGG [11] make it more suitable for stable mode-locking than Yb:CNGG with which we obtained 73 fs pulses using a SESAM, albeit with Ti:sapphire laser pumping [12].

Up to now, SWCNT-SA mode-locked Yb-lasers operating around 1 μm were usually pumped by Ti:sapphire lasers due to their good beam quality and high brightness: pulse

durations as short as 83 and 115 fs were obtained with different Yb:KY(WO₄)₂ and Yb:KLu(WO₄)₂ lasers, respectively [13,14]. However, the high cost and bulky volume of Ti:sapphire laser systems would limit the applications of such 1- μ m femtosecond sources and compact design would be impossible. Recently, SWCNT-SAs were used to mode-lock single-mode diode-pumped Nd:glass lasers [15] achieving 99 fs pulses at 1070 nm with an average output power of 10 mW.

In this work, we report first results on SWCNT-SA mode-locking of a diode-pumped Yb:CLNGG laser operating at \sim 1049 nm.

2. Experimental set-up

A transmission-type SWCNT-SA was employed in our experiment. Dried arc-discharge-made SWCNTs were dispersed in dichlorobenzene (DCB) via ultrasonic agitation, while PmPV was added to enhance their solubility. The SWCNT dispersion and a polymethyl methacrylate (PMMA) solution were then mixed in the same volume ratio. Finally, the SWCNT/PMMA mixture was deposited on a quartz substrate by spin coating and baked at 90°C. A more detailed description of the fabrication procedure can be found in [16]. The nonsaturable loss, modulation depth, saturation fluence and recovery time were measured to be 0.8%, 0.25%, 8.5 μ J/cm² and less than 1 ps, respectively.

The CLNGG crystal was doped with 5 at.% Yb. The crystal was grown by the conventional Czochralski method. The chemical formula for this disordered garnet can be expressed approximately as Yb:Ca₃Li_{0.26}Nb_{1.69}Ga_{3.05}O₁₂ (Yb:CLNGG). Spectroscopic data at 300 K for similar (4.3%) doping level measured by others [17] indicate a maximum absorption cross section of 2.18×10^{-20} cm² at 973 nm with a FWHM of 2.6 nm, and strongest emission band at 1026.5 nm with a maximum cross section of $\sim 2 \times 10^{-20}$ cm² and FWHM of \sim 20 nm. However, this band corresponds to transition to the second highest level of the ground-state multiplet while the transition to the highest level is seen as a shoulder at 1050 nm [17]. The upper state lifetime is known only for Yb:CNGG and amounts to 791 μ s [18].

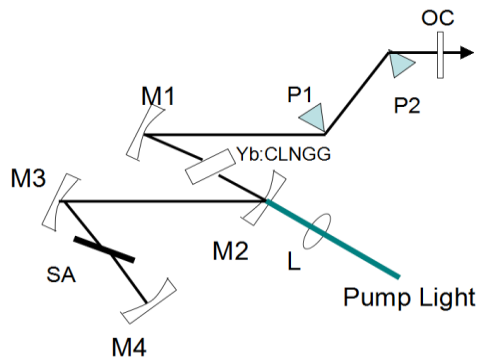


Fig. 1. Setup of the diode-pumped SWCNT-SA mode-locked Yb:CLNGG laser, M1-M4: concave mirrors (ROC = 100 mm); L: focusing lens; SA: SWCNT-SA; P1, P2: SF10 prisms, OC: output coupler.

An astigmatically compensated resonator with two folding mirrors (radius of curvature: ROC = 100 mm) was used for the laser, as shown in Fig. 1. The 2.18-mm-thick active element was oriented at Brewster angle and mounted without special water cooling. Two additional concave mirrors (ROC = 100 mm) separated by 150 mm created another cavity waist for the SWCNT-SA which was inclined under Brewster angle. The beam radii on the Yb:CLNGG crystal and SWCNT-SA were calculated using ABCD matrix formalism and amounted to 40 and 35 μ m, respectively. A distributed Bragg reflector (DBR) tapered diode laser with a nearly diffraction-limited beam quality and narrow spectral linewidth, operating at 979 nm was used as pump source [19]. The diode beam was focused by an $f = 6.28$ cm lens

through the folding mirror M2. The crystal absorption when the laser was interrupted amounted to 26%, independent of the pump power (shown in Fig. 2). This low value is due to the non-optimum pump wavelength, away from the absorption maximum. For the same reason, no absorption bleaching was observed, which differs from the behavior of Yb:CLNGG under Ti:sapphire pumping [12]. A pair of SF10 prisms with a separation of 54 cm was used for intracavity dispersion compensations and the laser was operated in the negative group velocity dispersion regime.

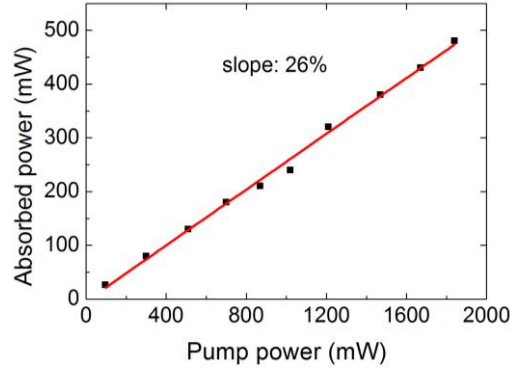


Fig. 2. Yb:CLNGG crystal absorption measured without lasing for the pump power available: experimental data (dots) and linear fit (solid line).

3. Results and discussion

To study the CW laser performance only the SWCNT-SA was removed from the beam path. Figure 3 shows the input-output characteristics of the CW Yb:CLNGG laser with different output couplers (OCs). A slope efficiency of 48% was achieved using a 5% OC. Thresholds as low as 85 mW (absorbed pump power) were obtained with the 0.4% OC. No signs of Kerr-lensing were observed when aligning the CW laser cavity.

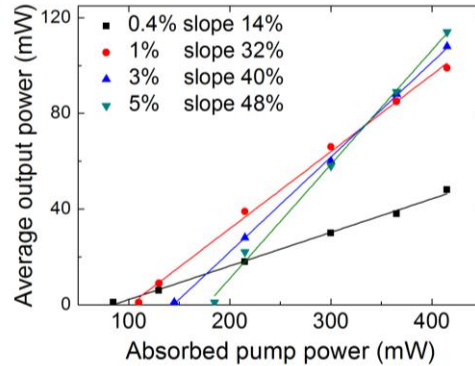


Fig. 3. Input-output characteristics of the diode-pumped CW Yb:CLNGG laser for different OCs.

With the SWCNT-SA inserted and careful realignment, mode-locking of the Yb:CLNGG laser was achieved with three OCs (0.4%, 1% and 3%). The shortest pulse duration of 90 fs (sech²-pulse shape) was obtained with the 0.4% OC. The output power was 20 mW in this case for an absorbed pump power of 365 mW. The calculated average fluence on the SWCNT-SA was about 1 mJ/cm², i.e. strong bleaching took place. Still, no damage or additional nonlinear losses occurred in this case. The corresponding optical spectrum was centered at ~1049 nm (transition to the highest level of the ground state multiplet) and had a FWHM of 14 nm, see Fig. 4 (a). This results in a time-bandwidth product of $\tau\Delta\nu = 0.345$,

slightly above the transform limit for sech^2 -shaped pulses. At higher pump powers the laser became unstable and a CW component appeared in the spectrum.

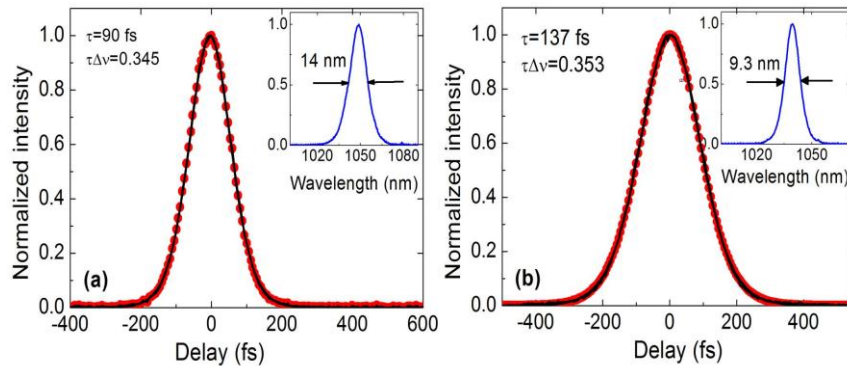


Fig. 4. Autocorrelation traces and spectra (insets) of the SWCNT-SA mode-locked Yb:CLNGG laser for 0.4% (a) and 3% (b) transmission of the output coupler. Red dots, measured autocorrelator data; solid curves, sech^2 -pulse shape fit.

The average output power increased to 90 mW at 410 mW of absorbed pump power (the maximum possible according to Fig. 3) when the 3% OC was employed, albeit at the expense of longer pulse duration, 137 fs using sech^2 -pulse shape for the fit. Figure 4 (b) shows the corresponding autocorrelation trace and spectrum measured. The generated pulses had a spectral width of 9.3 nm and the Fourier product remained almost the same, $\tau\Delta\nu = 0.353$. The increased inversion as a result of the higher OC transmission (cavity loss) shifted the central wavelength to 1039.3 nm where the gain is obviously higher in the three-level Yb-laser system. With an average power of 40 mW, pulse duration of 107 fs and Fourier product of 0.348, the 1% OC produced an intermediate result as could be expected.

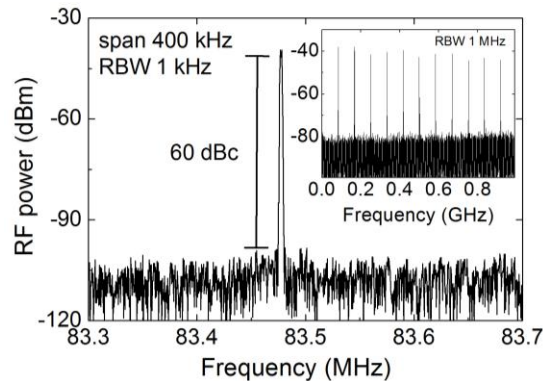


Fig. 5. Radio-frequency (RF) spectrum of the fundamental beat-note recorded with 1 kHz resolution and 1 GHz span (inset) for the mode-locked Yb:CLNGG laser with 0.4% OC.

From the radio frequency spectrum, see inset of Fig. 5, and the first beat note, recorded at 1 kHz resolution and shown in Fig. 5, no spurious modulations were visible down to 60 dBc, which indicates stable CW mode-locking of the Yb:CLNGG laser. The SWCNT-SA mode-locked laser was also self-starting.

The SWCNT-SA was translated along the cavity axis in order to vary the beam size but optimum operation was always in the second cavity waist.

As already mentioned, no mode-locking was achieved with the 5% OC. The stability criterion for CW mode-locking [20] requires, for the parameters of the present laser and assuming double action of the SWCNT-SA per round trip, i.e. a modulation depth of 0.5%, that the intracavity pulse energy exceeds ~ 60 nJ. Having in mind the possible effect of

inhomogeneous gain spectral broadening and soliton shaping, indeed the limit for stable CW mode-locking, comparing with Fig. 3, should lie between 3% and 5% output coupling. With higher output coupling such intracavity energy levels cannot be reached which explains the failure to achieve mode-locking with the 5% OC in the present experiment.

It should be mentioned that pulse durations down to 79 fs were obtained with the present set-up using the 0.4% OC but with compromised stability (sustained only for minutes and not reproducible).

A reflection-type SWCNT-SA was also tested in the experiment (substituting M4 in Fig. 1), which was manufactured by the same procedure as the transmission-type sample but on a high-reflectivity plane mirror. The shortest pulse durations with the 0.4% OC were again ~90 fs but there was always a CW component in the spectrum in this case. We attribute the inferior mode-locking performance with the reflection-type SWCNT-SA to its quality.

4. Conclusion

In conclusion, a diode-pumped Yb:CLNGG laser was mode-locked by a SWCNT-SA for the first time and the pulse duration of 90 fs obtained with 0.4% OC is the shortest, to the best of our knowledge for such type of laser. In fact we are not aware of any other diode-pumped solid-state laser based on a different ion and mode-locked by SWCNT-SA that produced shorter pulses apart from the very first demonstration with Er/Yb:glass operating at 1570 nm [4]. The maximum output power of the mode-locked Yb:CLNGG laser was 90 mW with a 3% OC at a repetition frequency of 83 MHz.

Since using SESAM in a Ti:sapphire laser pumped Yb:CNNG laser the durations were somewhat shorter, we plan to employ a SESAM as a SA also in the present diode-pumped Yb:CLNGG laser in the near future, for a direct comparison.

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