SWCNT-SA mode-locked Tm:$LuYO_3$ ceramic laser delivering 8-optical-cycle pulses at 2.05 µm

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Passive mode-locking of a Tm:$LuYO_3$ ceramic laser in the 2-µm spectral region is demonstrated by employing a SWCNT-SA. Almost chirp-free pulses as short as 57 fs, i.e., 8 optical cycles are produced at a repetition rate of 72.6 MHz (63 mW average power). The corresponding optical spectrum centered at 2045 nm, exhibits a bandwidth of 80 nm, which corresponds to almost chirp-free pulses. To the best of our knowledge, the results represent the shortest pulses generated by mode-locked Tm solid-state lasers. **© 2019 Optical Society of America**

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Thulium (Tm^{3+}) doped sesquioxide crystals are characterized by excellent thermo-optical and mechanical properties, relatively low maximum phonon energy and strong crystal fields. These features make them ideally suited for high power and broad-band tunable lasers in the 2-µm spectral region [1-4]. The strong crystal field splits the ground state of the Tm^{3+} ion up to 800 cm^{-1}, resulting in an emission bandwidth of more than 200 nm centered at 1950 nm [1]. Thus, the laser emission wavelength can easily be shifted to a wavelength above 2 µm, hence supporting stable femtosecond pulse generation even without purging the laser cavity. Using Tm-doped sesquioxide crystals, 175-fs pulse generation has been reported for a Tm:Lu_2O_3 laser mode-locked by a single-walled carbon nanotube (SWCNT) saturable absorber (SA) [5] and 166-fs pulses have been demonstrated for a Kerr-lens mode-locked Tm:Sc_2O_3 laser [6]. Further shortening of the pulse duration was realized by using mixed sesquioxides as gain media, resulting in compositional disorder, which provides slightly broader and smoother gain spectra as compared to the ordered sesquioxide crystals [1, 7, 8].

One notable disadvantage of the cubic sesquioxide crystals is their high melting temperature which exceeds 2400 □C, underlying a high mechanical and chemical stability, however, presenting a challenge for the crystal growth [1]. In comparison, sesquioxide ceramics, as an alternative solution, can be fabricated at much lower temperatures and without phase transition. The investigated Tm:$LuYO_3$ mixed ceramic was sintered by Hot Isostatic Pressing (HIP) method at a lower temperature of 1700 □C [9]. In fact, the performance of sesquioxide ceramics used as gain media for mode-locking is comparable or even superior to single crystals, e. g., 180-fs pulses were generated from a Tm:Lu_2O_3

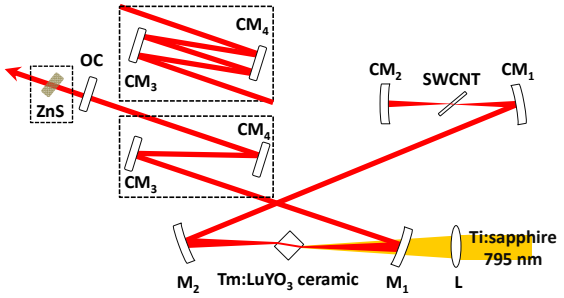


Fig. 1. Schematic of the SWCNT-SA mode-locked Tm:$LuYO_3$ ceramic laser. (L, lens; M_1-M_2, dichroic mirrors; CM_1-CM_4, chirped mirrors; OC, output coupler).

ceramic laser using semiconductor SA mirror (SESAM) for mode-locking [10], and pulses as short as 63 fs were reported recently employing a Tm:$(Lu_{2/3}Sc_{1/3})_2O_3$ mixed ceramic [11].

In this letter, mode-locking of a Tm:$LuYO_3$ mixed ceramic is demonstrated using a SWCNT-SA. After external dispersion management with a 3-mm-thick ZnS plate, pulses as short as 57 fs are achieved, i.e., 8 optical cycles estimated from the fringe-resolved interferometric autocorrelation.

The experimental setup of the SWCNT-SA mode-locked Tm:$LuYO_3$ ceramic laser is shown in Fig. 1. A standard X-shaped astigmatism compensated cavity was employed. The pump source was a continuous-wave (CW) Ti:sapphire laser tuned to 795 nm. Using a lens with a focal length of f = 70 mm, the pump beam was focused into the ceramic resulting in a beam radius of 30 µm. The mixed $(Lu_{0.5}Y_{0.5})_2O_3$ or $LuYO_3$ ceramic was doped with 3 at.% Tm^{3+} ions. The uncoated sample had dimensions of 3 × 3 × 3 mm^3. The ceramic was placed inside the cavity at Brewster's angle. The astigmatism was compensated by using two plano-concave mirrors, M_1 and M_2, both with radius of curvature, RoC = –100 mm. The calculated beam radius at the position of the Tm:$LuYO_3$ ceramic were 30 and 60 µm in the sagittal and tangential plane respectively. To mitigate the thermal load, the ceramic was wrapped with indium foil and tightly mounted in a copper holder which was water-cooled to 14.0 °C. The SWCNT-SA was deposited on a 1-mm-thick quartz substrate (~1% non-saturable loss, <0.5% modulation depth and <10 µJ/cm^2 saturation fluence around 2 µm [12]). It was placed in the second cavity waist under Brewster's angle, leading to beam radii of 70 and 105 µm in the sagittal and tangential plane, respectively. The second cavity waist was formed by two concave chirped mirrors, CM_1 and CM_2, with RoC = –100 and –50 mm respectively. Two further plane parallel chirped mirrors (CM_3 and CM_4) with 2 or 4 beam bounces were employed to optimize the dispersion. The group delay dispersion (GDD) provided by each of them was –125 fs^2 per bounce. Three plane-wedged mirrors with transmission of 3%, 1.5% and 0.5% were used as output couplers (OCs). A 3-mm-thick ZnS polycrystalline (ceramic) with group velocity dispersion (GVD) ~ +155 fs^2/mm at 2.05 µm was employed for extracavity dispersion optimization. A commercial autocorrelator (pulseCheck, APE GmbH, Berlin, Germany) and a home-built second-harmonic generation (SHG) frequency-resolved optical gating (FROG) apparatus were used for temporal characterization of the output.

At first the CW wavelength tunability of the Tm:$LuYO_3$ ceramic laser was investigated by using a 3.2-mm-thick birefringent quartz plate (Lyot filter) in the cavity (without chirped mirrors and SWCNT-SA). The Tm:$LuYO_3$ ceramic pump absorption showed

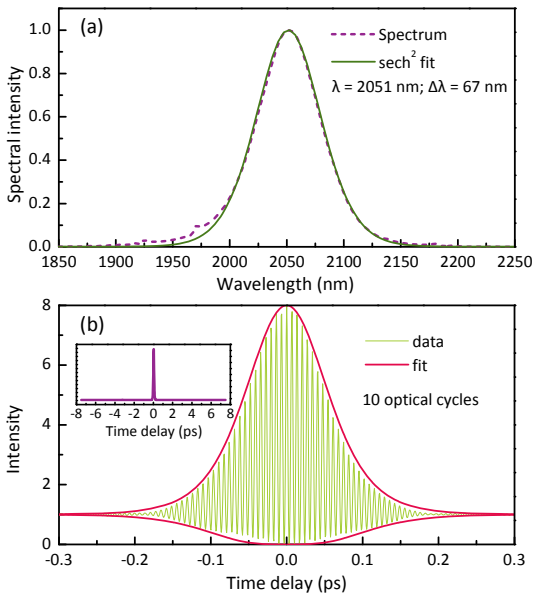


Fig. 2. Mode-locked Tm:$LuYO_3$ ceramic laser (T_{OC} = 3%). Optical spectrum (a) and interferometric autocorrelation trace (b). Inset (b): corresponding noncollinear autocorrelation trace on a time scale of ±7.5 ps.

bleaching effects and depended on the specific OC used: e.g., it amounted to □63% under lasing conditions with T_{OC} = 0.5%. For this OC, the absorbed power at threshold was as low as □140 mW. Using T_{OC} = 1.5%, the laser was tunable from 1909 to 2109 nm. The maximum output power obtained with this output coupler amounted to 440 mW at 2074.2 nm, which coincided with the free-running wavelength in this case. This 200 nm CW tuning range exceeds the one reported for the Tm:$(Lu_{2/3}Sc_{1/3})_2O_3$ mixed ceramic laser of 130 nm, achieved under similar conditions [11].

For the mode-locking experiments, the SWCNT-SA was inserted in the cavity. The physical cavity length was extended by introducing the chirped mirrors, initially to ~1.75 m using two beam bounces per round trip on each of the mirrors CM_3 and CM_4. In this configuration the total round-trip GDD amounted to –1100 fs^2 taking into account also the material dispersion mainly due to the 1-mm-thick quartz substrate of the SA (GVD: –112 fs^2/mm at 2050 nm). The contribution of the ceramic sample, estimated by averaging the corresponding refractive index data for Y_2O_3 and Lu_2O_3 crystals [13], was roughly +50 fs^2 per round trip and obviously negligible.

Mode locking of the Tm:$LuYO_3$ ceramic laser was self-starting for all output couplers applied. Using T_{OC} = 3% an average output power of 210 mW was achieved for 2.1 W absorbed pump power. Taking into account the repetition rate of the laser, 85.6 MHz, the pulse energy amounts to ~2.5 nJ. Thus the average pulse fluence on the SWCNT-SA was calculated to be ~350 µJ/cm^2. Figure 2(a) shows the corresponding optical spectrum with a central wavelength at 2051 nm. The measured spectral profile can be fitted with a $sech^2$-function, giving a FWHM of 67 nm. The interferometric autocorrelation trace is shown in Fig. 2(b), assuming a $sech^2$ pulse shape, yielding a pulse duration (FWHM) of 68 fs and a time-bandwidth product (TBP) of 0.325. The analysis of the autocorrelation trace envelopes indicates nearly chirp-free pulses [14] and counting the fringes one arrives at 10 optical cycles for the pulse duration. Single pulse operation without satellites

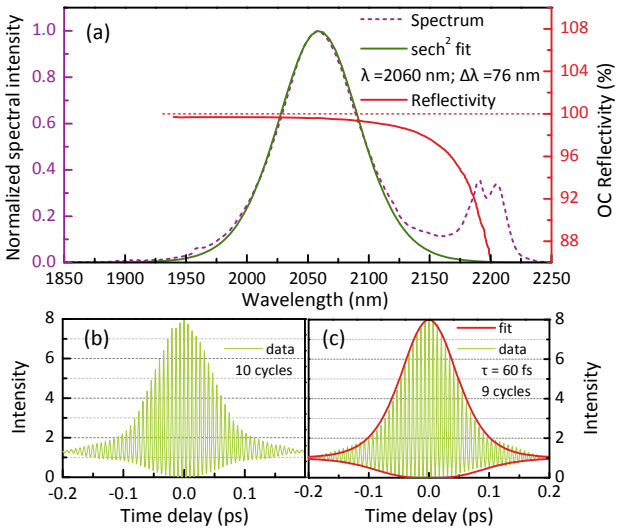


Fig. 3. Optical spectrum (a) of the mode-locked Tm:$LuYO_3$ ceramic laser with T_{OC} = 0.5%, and the corresponding interferometric autocorrelation traces before (b) and after (c) compression. The red curve in (a) shows the reflectivity of the output coupler.

was confirmed by recording the noncollinear autocorrelation trace on a longer time scale of ±7.5 ps, see the inset of Fig. 2(b). It should be noted that the on-axis intracavity laser intensity on the ceramic was estimated to be 43 GW/cm², which is comparable to the 45 GW/cm² reported for the Kerr-lens mode-locked Tm:Sc_2O_3 laser in [6], delivering 298-fs pulses. However, pure Kerr-lens mode-locking was not achieved in our laser configuration.

The present results seem of high importance for Ho-doped power amplifiers, mainly based on Ho:YLF or Ho:YAG [15] because the number of 2-μm laser sources which meet the spectral requirements for seeding these amplifiers is very limited [16]. The center wavelength of the mode-locked Tm:$LuYO_3$ ceramic laser of 2051 nm exactly fits the gain maximum of Ho:YLF.

Subsequently, an OC with transmission of 0.5% was used in the mode-locked Tm:$LuYO_3$ mixed ceramic laser, delivering 70 mW average output power at 2 W of absorbed pump power. Compared to the laser with T_{OC} = 3% the average pulse fluence on the SWCNT-SA increased to 700 μJ/cm². As shown in Fig. 3(a), the central emission wavelength was red-shifted to 2060 nm. This is attributed to the lower population inversion required thereby resulting in stronger reabsorption during the lasing process. As in the case of the operation with T_{OC} = 3%, the spectral envelope was fitted well with a sech²-intensity profile except for the small peaks in the long-wave wing, which originated from the leakage of the 0.5% OC. (see its reflectivity curve in Fig. 3(a)). The pulse spectral FWHM amounted to 76 nm. The measured pulse duration using noncollinear autocorrelation amounted to 67 fs. In this case, the on-axis intracavity intensity on the ceramic reached 86 GW/cm², leading to a further spectral broadening due to a stronger self-phase modulation (SPM) compared to the T_{OC} = 3% case. However, the increased TBP value of 0.36 indicated the existence of a residual pulse chirp. This chirp could be eliminated using the 3-mm-thick ZnS ceramic sample. Interferometric autocorrelation traces recorded without and with the ZnS sample are shown in Fig. 3(b) and 3(c), respectively. Obviously, the pulse duration was reduced to 60 fs, i.e., from 10 to 9 optical cycles. The fits to the optical spectrum and the envelopes of the interferometric

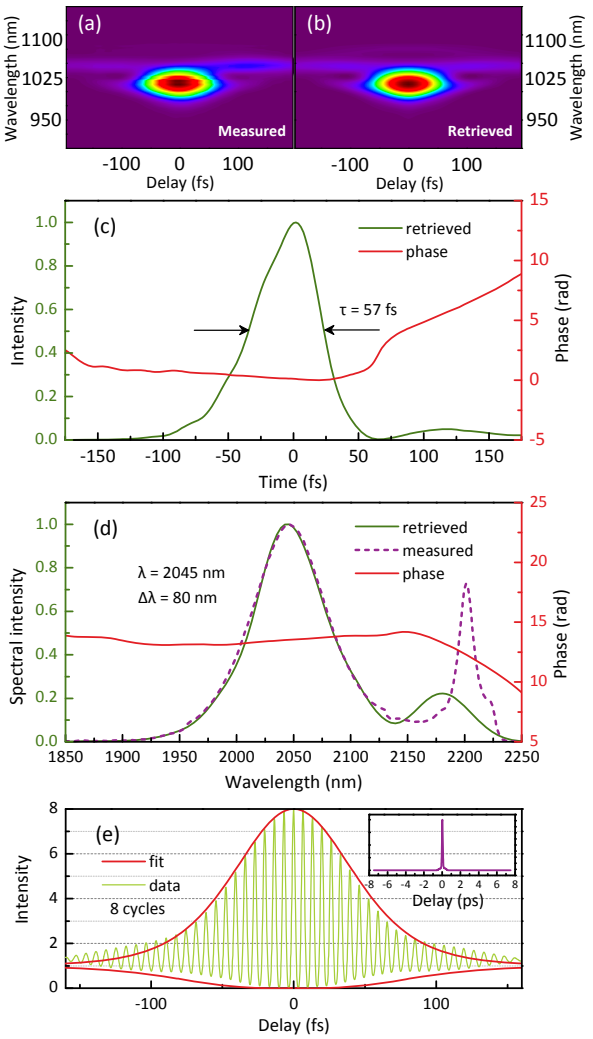


Fig. 4. Mode-locked Tm:$LuYO_3$ ceramic laser (T_{OC} = 0.5%) – shortest pulses. Measured (a) and retrieved (b) SHG-FROG traces after external pulse compression using ZnS, and the reconstructed temporal (c) and spectral (d) intensity and phase profiles; the dashed line in (d) is the measured optical spectrum. (e) interferometric autocorrelation trace. Inset (e): corresponding noncollinear autocorrelation trace on a time scale of ±7.5 ps.

autocorrelation assuming a sech² pulse shape in Fig. 3, resulted in a TBP of 0.322.

The shortest pulses were generated by lengthening the cavity with the 0.5% OC up to ~2 m in order to further increase the intracavity pulse energy. Despite the slight drop in the average output power down to 63 mW, the average intracavity fluence on the SWCNT-SA increased to ~750 μJ/cm². The stronger SPM was balanced by increased GDD = –1600 fs² realized by four bounces per round trip on each of the mirrors CM_3 and CM_4, see Fig. 1. The output pulses behind the ZnS plate were characterized by SHG-FROG.

Figures 4(a)–4(d) display the measured and reconstructed SHG-FROG traces together with the retrieved temporal and spectral profiles and phases. The FROG error between the measured and reconstructed SHG-FROG traces on a 128 × 128 grid was 0.006. As shown in Fig. 4(c), the retrieved pulse duration amounted to 57 fs, corresponding to ~8 optical cycles. From Fig. 4(d), it can be seen

that the reconstructed spectrum profile coincided well with the measured one, except for the already discussed sideband at longer wavelengths (see Fig. 3(a)). The center wavelength was at 2045 nm and the spectral FWHM amounted to 80 nm, giving a TBP of 0.327 which indicates almost transform-limited pulses. The residual chirp was deduced to be ±20 fs^2 by fitting the phase profile of the main spectral peak. At such a low chirp value, it was impossible to determine unambiguously the sign with the help of a substrate. The 8 optical cycles under the pulse envelope were confirmed by the recorded fringe-resolved interferometric autocorrelation trace (Fig. 4(e)). Single pulse operation without satellites was confirmed by a measurement on a longer time scale (inset in Fig. 4(e)). Since the pulses were almost chirp-free, the deviation from the fit based on a $sech^2$-intensity pulse shape was again attributed to the long-wavelength leakage of the 0.5% OC.

Note that the ZnS plate used outside the cavity in the present set-up operates only as a linear compression element which in fact helps to finely adjust the net intracavity GDD that can be controlled only in a stepwise manner by the chirped mirrors. The estimated nonlinear phase shift (B-integral) for the axial field does not exceed 0.2 mrad according to the nonlinear refractive index of ZnS near 2-μm [17]. As expected for such small values, no difference in the output spectrum with and without the ZnS plate could be observed.

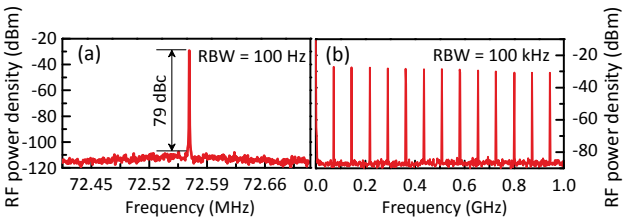


Fig. 5. Radio frequency spectra of the mode-locked Tm:$LuYO_3$ ceramic laser in 300 MHz (a) and 1 GHz (b) span range (T_{OC} = 0.5%).

To assess the stability of the SWCNT-SA mode-locked Tm:$LuYO_3$ ceramic laser, radio frequency (RF) spectra were recorded for the shortest pulses. Figure 5(a) shows the fundamental beat note at 72.57 MHz with an extinction ratio above the noise level of 79 dBc. This high contrast and the uniform harmonic beat notes in a wide span of 1 GHz (see Fig. 5(b)) indicate stable and CW mode-locked operation. All presented mode-locked Tm:$LuYO_3$ ceramic laser configurations run stable for hours.

In conclusion, we have demonstrated a SWCNT-SA mode-locked Tm:$LuYO_3$ ceramic laser generating sub-10-optical-cycle pulse in the 2-μm spectral range. Its free-running laser wavelength above 2 μm together with a 200 nm broad CW laser tuning range underline the excellent suitability of this novel Tm:$LuYO_3$ mixed ceramic for sub-100-fs pulse generation. In the mode-locked regime a maximum output power of 210 mW was achieved at 85.6 MHz repetition rate. In this case 68 fs pulses were generated with an optical spectrum centered at 2051 nm. Broadening of the spectral FWHM from 67 to 80 nm was achieved by increasing the intracavity power when using a low transmission of the OC (T_{OC} = 0.5%). Almost chirp-free pulses as short as 57 fs (TBP = 0.327) at 2045 nm, with an output power of 63 mW were achieved, corresponding to only 8 optical cycles, which to the best of our knowledge is the shortest pulse duration ever reported for a mode-locked Tm laser in the 2 μm spectral range. Although pure Kerr-lens mode locking was not observed with the present set-up, from estimations of Tm:Lu_2O_3 ceramic at 2070 nm [10], the nonlinear refractive index shall be of the order of 3.3×10^{-16} cm^2/W, i.e., rather close to the value for Ti:sapphire near 800 nm, and we expect such a regime to be more easily achievable with somewhat longer ceramic samples of Tm.$LuYO_3$.

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