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SESAM mode-locked Tm:LuYO₃ ceramic laser generating 54-fs pulses at 2048 nm

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Mode-locked laser operation near 2.05 μm based on a mixed sesquioxide Tm:LuYO₃ ceramic is demonstrated. Continuous-wave and wavelength tunable operation is also investigated. Employing a GaSb-based SESAM as a saturable absorber, a maximum average output power of 133 mW is obtained for a pulse duration of 59 fs. Pulses as short as 54 fs, i.e., 8 optical cycles are generated at a repetition rate of ~ 78 MHz with an average output power of 51 mW. To the best of our knowledge, this result represents the shortest pulse duration ever achieved from Tm-based solid-state mode-locked lasers.

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Thulium (Tm)-doped laser materials feature the advantages of broadband gain spectra due to the $^3\text{F}_4 \rightarrow ^3\text{H}_6$ electronic transition of Tm^{3+} , power scaling capabilities with commercially available, AlGaAs pump laser diodes at ~ 800 nm,

and high quantum efficiency via a one-for-two cross-relaxation process. These features are attractive in particular for the development of broadband tunable and femtosecond (fs) solid-state lasers in the 2- μm spectral range. Nevertheless, sub-100-fs mode-locked (ML) bulk lasers were not reported until 2017: Using graphene as a saturable absorber (SA), the monoclinic Tm:MgWO₄ crystal delivered 86-fs pulses at 2017 nm [1]. Subsequently, single-walled carbon nanotube-based SA (SWCNT-SA) ML lasers using Tm-doped disordered garnets also produced sub-100-fs pulse durations at ~ 2 μm [2,3].

Among the Tm^{3+} -doped laser hosts, the cubic (bixbyite structure, space group $Ia\bar{3}$) sesquioxides RE₂O₃, where RE = Lu, Y and Sc, are of particular interest for sub-100-fs ~ 2 - μm solid-state lasers because of their extremely broad and flat gain spectra extending up to ~ 2.1 μm due to the strong Stark splitting of the ground state of Tm^{3+} ($^3\text{H}_6$, > 800 cm^{-1}) in combination with strong electron-phonon interaction and their high thermal conductivity [4]. The first sub-100-fs pulses from a Tm^{3+} -doped crystalline sesquioxide laser have been demonstrated with a Kerr-lens ML Tm:Sc₂O₃ laser at 2108 nm [5]. However, the optical quality remains a critical factor for such crystals due to the very high melting point of the RE₂O₃ compounds (typically

2400 - 2500 °C) and possible structural phase transitions in some of them upon cooling [4]. In contrast, ceramic fabrication circumvents such problems due to the relatively low sintering temperature [6]. Moreover, it is easier to mix isostructural sesquioxide ceramics leading to compositional disorder and broader absorption and gain spectra, the latter being beneficial for the generation of even shorter laser pulses. Sub-10 optical-cycle (63-fs) pulses at 2057 nm were first achieved with a mixed Tm:(Lu,Sc)₂O₃ ceramic laser using a Semiconductor Saturable Absorber Mirror (SESAM) for mode-locking [7]. More recently, pulses as short as 57 fs at 2045 nm were reported for a mixed Tm:LuYO₃ ceramic laser ML by a SWCNT-SA [8].

The promising spectroscopic features and the previous ML laser results motivated us to further explore the potential of the mixed Tm:(Lu_{0.5}Y_{0.5})₂O₃ or Tm:LuYO₃ ceramic. This ceramic is synthesized by the hot isostatic pressing (HIP) method at 1700 °C. It exhibits a rather broad absorption band of 26 nm at full width at half-maximum (FWHM) centered at 796.5 nm [9], which in fact is a combination of few narrower lines, similar to the composite spectra of Tm:Lu₂O₃ and Tm:Y₂O₃ [10]. In the present work, a cubic shape, 3-mm-thick, uncoated Tm:LuYO₃ ceramic sample was used with 3 at.% Tm³⁺-doping (measured small-signal absorption: 66%).

Nowadays SESAMs are the most widely used SAs for ML bulk solid-state lasers that ensure stable and self-starting operation. However, their availability for specific wavelength ranges is limited, in particular around 2 μm. Recent development of SESAM for the 2-μm spectral range has focused on InGaAsSb quantum-wells (QWs) embedded in GaSb in which offset and band-gap can be tailored to encompass a spectral range across 1.9 – 3.0 μm. Such 2-μm SESAMs show a fast (few ps) absorption recovery time in particular for the near-surface design [11] and an exceptionally broad reflection band of ~300 nm due to the use of lattice-matched GaSb/AlAsSb distributed Bragg reflectors (DBRs) [12]. Sub-100-fs pulse ML operation of Tm³⁺ doped or Tm³⁺, Ho³⁺ co-doped laser materials at ~2 μm using such SESAMs has already been demonstrated [13]. In this work, we focus on the study of the mixed Tm:LuYO₃ ceramic as an active medium in combination with a GaSb-based SESAM for ML operation. Nearly transform-limited pulses as short as 54 fs are reported, the shortest pulse duration ever achieved for any Tm-based solid-state laser. Additionally, continuous-wave (CW) tunable laser operation from 1909 to 2109 nm is characterized.

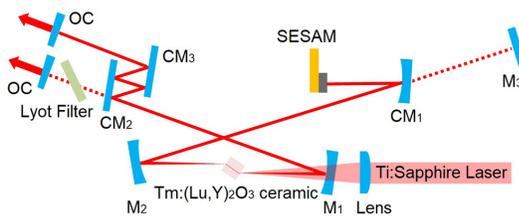


Fig. 1. Setup of the ML Tm:LuYO₃ laser: M₁-M₂: concave folding mirrors (RoC = 100 mm), M₃: rear mirror in CW operation; CM₁-CM₃, chiral mirrors; OC: output coupler.

The experimental setup is shown in Fig. 1. The ceramic sample was mounted on a water-cooled copper block (coolant temperature: 14 °C) and placed at Brewster's angle between

two dichroic folding mirrors M₁ and M₂. The pump source was a CW narrow-linewidth Ti:Sapphire laser tuned to 795.3 nm. It was focused into the ceramic sample with an $f = 70$ mm lens to a beam radius of 30 μm. The CW laser performance of the mixed Tm:LuYO₃ ceramic was studied with a standard 4-mirror astigmatically compensated X-type cavity including a plane rear reflector M₃ and a plane-wedged output coupler (OC). The laser cavity mode inside the ceramic sample was estimated using the ABCD formalism giving a waist radius of 30 and 76 μm in the sagittal and tangential planes, respectively.

As can be seen from Fig. 2(a), in the CW laser regime, a maximum output power of 603 mW was obtained for 3% OC transmission at an absorbed pump power of 2.12 W corresponding to a slope efficiency of 33.2%. The single pass pump absorption under lasing conditions depended on the OC and amounted to 62.2% for this OC, revealing some absorption bleaching. The central wavelength changed with T_{OC} from 2060 to 2076 nm, Fig. 2(b).

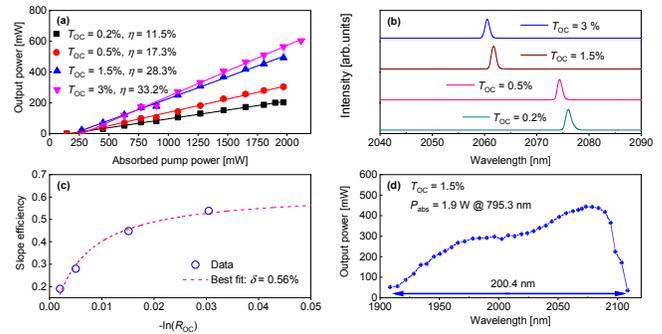


Fig. 2. CW laser performance of the mixed Tm:LuY₂O₃ ceramic for different OCs (a); laser spectra (b); slope efficiency as a function of the OC reflectivity $R_{OC}=1-T_{OC}$ (c); and spectral tuning range obtained with a Lyot filter (d).

The total round-trip resonator losses δ (reabsorption losses excluded) were estimated with the Caird analysis by fitting the measured slope efficiency as a function of the OC reflectivity $1-T_{OC}$ [14]. The best fit gave $\delta = 0.56\%$, Fig. 2(c). A Lyot filter (3.2-mm-thick quartz birefringent plate) was placed at Brewster's angle close to the OC for wavelength tuning. With the 1.5% OC, an extremely broad tuning range of >200 nm was achieved as shown in Fig. 2(d), limited by the free spectral range of the filter and the reflectivity band of the cavity mirrors. It exceeded the tuning range reported for the Tm:(Lu,Sc)₂O₃ ceramic under similar conditions by roughly 70 nm [7].

For ML operation, M₃ was replaced by a curved chiral mirror CM₁ with RoC = 100 mm in order to create a second intracavity beam waist on the SESAM to increase the fluence for efficient bleaching of the SA. The calculated radius of this beam waist was 120 μm. The GaSb-based SESAM applied in this experiment was analogous to Sample 7, presented in Table 3 of [11]. Thus it contained two InGaAsSb QWs with a 10 nm barrier placed below the surface (50 nm GaSb cap) which was uncoated. The photoluminescence of the present sample was centered at 2090 nm. For such structures we measured typical recovery times with a fast and a slow component of about 240 fs and 21 ps, respectively [11], however, the saturable/non-

saturable losses and the saturation fluence are still difficult to evaluate.

The intracavity group delay dispersion (GDD) was optimized by two extra plane CMs (CM₂ and CM₃) in the other cavity arm, changing the number of bounces. All CMs had a GDD of -125 fs² per bounce. Applying 3, 5 and 7 bounces (single pass) on the plane CMs, the total round-trip GDD amounted to -887, -1387 and -1887 fs², respectively (including the contribution of the ceramic sample: -137 fs², estimated by averaging the refractive index for Y₂O₃ and Lu₂O₃ [15]). For three of the OCs ($T_{OC} = 1.5\%$, 0.5% and 0.2%), stable and self-starting sub-100-fs pulse generation was obtained, however, only for 5 bounces on the CMs.

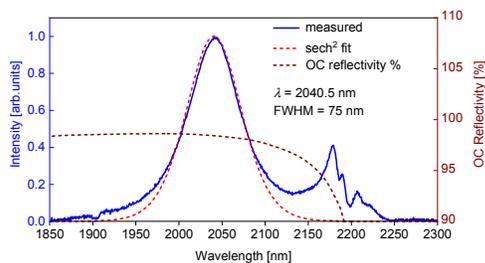


Fig. 3. Spectrum of the ML Tm:LuY₂O₃ ceramic laser using $T_{OC}=1.5\%$ (left axis) and OC reflectivity $R_{OC}=1 - T_{OC}$ (right axis).

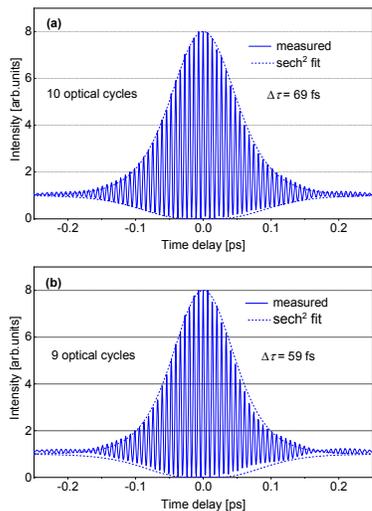


Fig. 4. Interferometric autocorrelation traces before (a) and after (b) external compression for $T_{OC} = 1.5\%$. $\Delta\tau$: fitted pulse duration (FWHM).

Based on this cavity configuration, pulses as short as 69 fs with an average output power of 133 mW (for an absorbed pump power of 1.68 W) were obtained directly from the ML laser with $T_{OC}=1.5\%$. The geometrical cavity length was ~ 1.92 m which resulted in a pulse repetition rate of ~ 78 MHz. The spectrum of the ML laser, recorded with a resolution of 0.5 nm, was centered at 2040.5 nm and had a sech²-fitted FWHM of 75 nm, as shown in Fig. 3. The observed satellite at longer wavelengths is associated with leakage through the OC whose transmission drastically increases at ~ 2200 nm. In fact, it

does not present an artefact but is a kind of pulse shaping mechanism which alleviates a trend towards multi-pulse instability in the ML regime due to excessive intracavity energy for the given negative GDD. It could be ideally suppressed in future experiments by optimized characteristics of the OC. However, achieving yet broader reflectivity bands for such an OC, is indeed a technological challenge.

The envelopes of the recorded interferometric autocorrelation trace in Fig. 4(a) were well fitted assuming a sech²-shaped pulse with a duration of 69 fs (FWHM), equivalent to 10 optical cycles. The resulting time-bandwidth product amounted to 0.373 indicating some residual chirp. The latter was eliminated by additional external compression using a 3-mm-thick ZnS polycrystalline plate. A pulse duration as short as 59 fs was achieved, confirmed by the measured interferometric autocorrelation trace in Fig. 4(b), representing 9-optical-cycle pulses. Due to the unaltered spectrum, the time-bandwidth product dropped to 0.319 indicating almost bandwidth limited pulses.

The pulse duration was further shortened by reducing the OC transmission to 0.5% at the expense of the average output power. Thus, the shortest pulses directly generated from the self-starting ML laser had a FWHM duration of 66 fs. At ultimate stability, the average output power amounted to 51 mW for 1.5 W of absorbed pump power. The corresponding spectrum was centered at 2048 nm with a sech²-fitted FWHM of 82 nm. The slightly chirped pulses with a time-bandwidth product of 0.387 were externally compressed with the same 3-mm-thick ZnS plate. Their characterization included interferometric autocorrelation and SHG-FROG (second-harmonic generation frequency-resolved optical gating) with a 1-mm-thick type-I LiNbO₃ crystal. Assuming a sech² pulse shape, the autocorrelation measurement (Fig. 5) gave a pulse duration of 54 fs representing 8 optical cycles by counting the fringes. The absence of post- or pre-pulses was confirmed by a measurement on a 15-ps time span (inset Fig. 5).

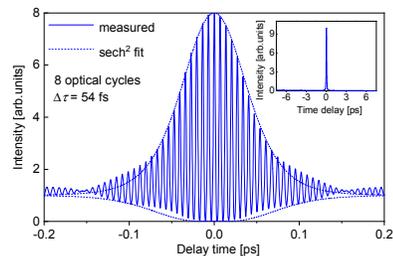


Fig. 5. Interferometric autocorrelation trace of the shortest pulses from the ML Tm:LuYO₃ ceramic laser obtained with $T_{OC} = 0.5\%$. The fit assumes a sech²-pulse shape. Inset: noncollinear autocorrelation trace on a time span of 15 ps.

The FROG results are summarized in Fig. 6. The measured FROG spectrogram was reconstructed with an error of 0.0048 on a 128×128 grid, Fig. 6(a) and (b). The retrieved FROG spectrum with a FWHM of 82 nm, centered at 2048 nm, is shown in Fig. 6(c). It coincides almost perfectly with the one independently measured with a spectrometer, except for the small satellite at longer wavelengths. A pulse duration of 55 fs was derived from the FROG retrieval as shown in Fig. 6(d), in excellent agreement with the independently measured

autocorrelation trace; cf. Fig. 5. The residual linear chirp parameter was derived from a parabolic fit to the spectral phase leading to ± 100 fs². It was not possible to determine the sign with a suitable substrate because of the small chirp value. The time-bandwidth product after compression (0.316) was indicative of Fourier-limited pulses.

Compared to the experimental conditions in [8], stronger self-phase modulation can be expected in the present work due to the higher intracavity intensity in the ceramic sample (~ 50 GW/cm² on-axis for the shortest pulses). The same is true for the spatial Kerr-lens effect. Concerning the origin and role of the spectral satellite at longer wavelengths, cf. Fig. 3 and Fig. 6(c), it shall be emphasized that the drop in the OC reflectivity is associated with abrupt variation of its GDD. However, the same is true also for the CMs around 2200 nm. In fact, from experiments using a different OC design, which ensured extended reflectivity at longer wavelengths, it seems that a similar, though much weaker in amplitude satellite is still present, and thus its origin might be related to higher order dispersion near 2200 nm. However, a stronger satellite occurring for an OC with a narrower reflection band as the one employed in the present work seems to stabilize the soliton regime in this laser where the intracavity GDD can be optimized only in a discrete manner, ultimately contributing to the achievement of the shortest pulse duration.

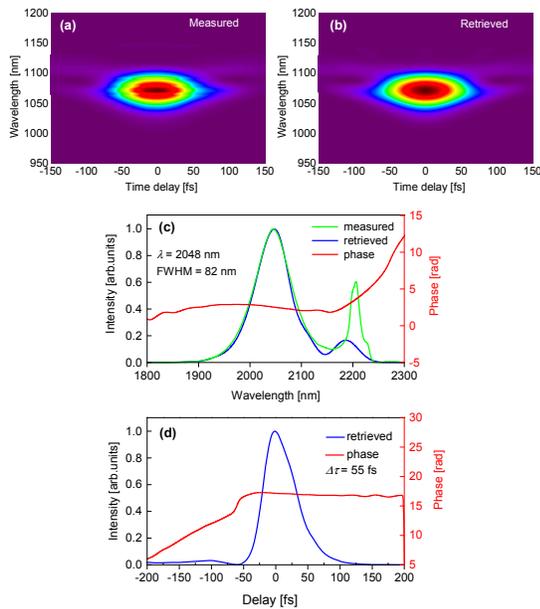


Fig. 6. SHG FROG characterization for the shortest pulses of the Tm:LuYO₃ ceramic laser ($T_{oc} = 0.5\%$): Measured (a) and retrieved (b) FROG traces; retrieved and measured spectra, and retrieved spectral phase (c); retrieved temporal intensity profile and phase (d).

Figure 7 shows the measured radio frequency (RF) spectra of the fundamental beat note at ~ 78 MHz with a resolution bandwidth (RBW) of 300 Hz and a 1-GHz-wide span (RBW: 10 kHz) to verify the stability of the ML laser for the shortest pulse operation. The very high extinction of 76 dBc and the absence of any spurious modulation are evidences for stable CW-ML operation of the mixed Tm:LuYO₃ ceramic laser.

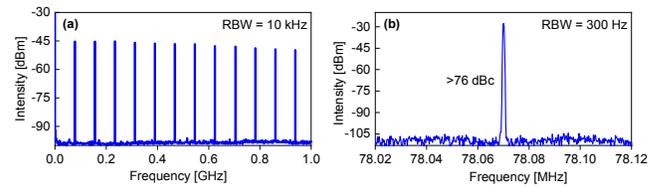


Fig. 7. Radio frequency spectra of the ML mixed Tm:LuYO₃ sesquioxide ceramic laser: 1-GHz span (a) and fundamental beat note (b).

In conclusion, we present a detailed investigation of CW, tunable and mode-locked laser operation of a mixed sesquioxide Tm:LuYO₃ ceramic laser at ~ 2 μ m. Using the same gain medium and GDD compensation scheme, it was possible to directly compare mode-locking initiated by a SESAM and a SWCNT-SA [8]. The pulse duration of 54 fs achieved in the present work by using a GaSb-based SESAM as a SA, represents the shortest pulse ever reported for any Tm-based bulk solid-state laser. It demonstrates that a resonant-type SA can be equally good or even better for mode-locking of such femtosecond lasers. Whereas from previous studies it is known that such SESAMs typically show slower response, larger modulation depths and lower non-saturable losses compared to SWCNT-SAs [13], the present results indicate that these characteristic parameters are not of crucial importance and can vary in quite broad ranges, see Table 1 in [13]. Finally, we note that while Tm-fiber lasers have produced slightly shorter pulse durations, this was only the case after external compression in a prism pair [16] while the oscillator itself operated in the chirped pulse regime, delivering pulse durations of the order of 1 ps.

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Disclosures. The authors declare no conflicts of interest.

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