







SESAM mode-locked Tm:Y₂O₃ ceramic laser

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Abstract: We demonstrate a widely tunable and passively mode-locked Tm:Y₂O₃ ceramic laser in-band pumped by a 1627-nm Raman fiber laser. A tuning range of 318 nm, from 1833 to 2151 nm, is obtained in the continuous-wave regime. The SESAM mode-locked laser produces Fourier-transform-limited pulses as short as 75 fs at ~ 2.06 μm with an average output power of 0.26 W at 86.3 MHz. For longer pulse durations of 178 fs, an average power of 0.59 W is achieved with a laser efficiency of 29%. This is, to the best of our knowledge, the first mode-locked Tm:Y₂O₃ laser in the femtosecond regime. The spectroscopic properties and laser performance confirm that Tm:Y₂O₃ transparent ceramics are a promising gain material for ultrafast lasers at 2 μm.

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1. Introduction

Cubic sesquioxides (RE₂O₃, where RE = Lu, Y or Sc) exhibit excellent chemical stability and superior thermal-mechanical properties which is partially related to their high melting temperature of more than 2400 °C [1]. In combination with the strong crystal fields that can result in broad and smooth gain spectra of the active dopant ions [2], they represent an attractive class of materials both for high average power (kilowatt [3]) and ultrafast (few-optical-cycle pulses [4]) lasers. In addition, the relatively low maximum phonon energy of the order of 600 cm⁻¹, makes them suitable also for operation at longer wavelengths, up to the 3-μm upper limit for oxide type hosts [1]. When doped with thulium ions (Tm³⁺), the strong splitting of the ground-state multiplet (³H₆) enables laser emission wavelength exceeding 2 μm, thus avoiding the structured water vapor air absorption and supporting stable ultrashort pulse generation [2]. To date, 175-fs pulses were reported from a single-walled carbon nanotubes based saturable absorber mode-locked Tm:Lu₂O₃ laser [5]. With a high quality-factor cavity, a Kerr-lens mode-locked Tm:Sc₂O₃ laser delivered 72-fs pulses at 2.1 μm [6]. Most recently, sub-60-fs pulses were generated by using

compositionally disordered "mixed" sesquioxide ceramics [7,8] or by combining the gain spectra of two different materials, i.e., Tm:Lu₂O₃ and Tm:Sc₂O₃ [9]. Tm:Y₂O₃ has been less studied due to the phase transition in Y₂O₃ close to the melting point which is expected to affect the optical quality. However, the segregation of Tm ions in Y₂O₃ should be similar to Lu₂O₃ and Y₂O₃ also exhibits the lowest maximum phonon energy [1]. Mode-locking with Tm:Y₂O₃ has been previously reported only in the picosecond regime [4].

The impressive results obtained with other Tm-doped sesquioxide materials motivated us to study femtosecond mode-locked operation of the Tm:Y₂O₃ laser. Compared to the conventional growth of high-optical-quality crystalline sesquioxides [1,2], polycrystalline transparent ceramics exhibit a much lower sintering temperature which makes them feasible for large-size fabrication with good optical quality [10]. In the present work, we fabricated a high-optical-quality Tm:Y₂O₃ transparent ceramic by using the hot isostatic pressing method. The Tm:Y₂O₃ ceramic laser was pumped by a 1627-nm Raman fiber laser and mode-locked in the femtosecond regime by employing a semiconductor saturable absorber mirror (SESAM). The high brightness of the single-mode fiber laser together with the high quantum efficiency (> 77%) of such in-band pumping into the upper laser level offer specific advantages for sub-100-fs pulse generation with a relatively high efficiency.

2. Fabrication and spectroscopic properties of Tm:Y₂O₃ ceramics

Y₂O₃ transparent ceramics doped with 2 at.% Tm³⁺ were fabricated with vacuum sintering and hot-isostatic pressing methods. The Tm:Y₂O₃ raw powders were synthesized by using the precipitation method; details regarding the yttria powder synthesis can be found in a previous report [11]. Next, the green bodies were prepared by pressing the raw powders in a stainless steel die (22 mm in diameter) with a pressure of about 10 MPa, and thereafter further compressed with a cold isostatic press (CIP) under 200 MPa. Subsequently, the shaped samples were sintered in a vacuum sintering furnace equipped with a tungsten mesh heating element. The sintering took place for 7 h at 1500 °C under a low vacuum of less than 10⁻³ Pa. To achieve ultimate densification, the samples were further hot-isostatically pressed (HIPed) at 1450 °C and 196-MPa Ar gas pressure for 3 h. Finally, the fabricated Tm:Y₂O₃ ceramic samples were annealed at 1000 °C for 10 h under normal atmospheric conditions.

A polished 2-mm-thick Tm:Y₂O₃ ceramic sample was used for the measurement of absorption spectrum at room temperature. With the concentration of the Tm³⁺ ions ($N_{\text{Tm}} = 5.26 \times 10^{20}$ cm³), the absorption cross-section (σ_{abs}) for the ³H₆ → ³F₄ transition was calculated as shown in Fig. 1(a). The maximum σ_{abs} amounts to 7.47×10^{-21} cm² at 1635 nm, which is exactly located in the spectral region of the wavelength-tunable Raman-shifted Er-fiber laser, and thus suitable for the in-band pumping by using such a light source [12]. The stimulated emission

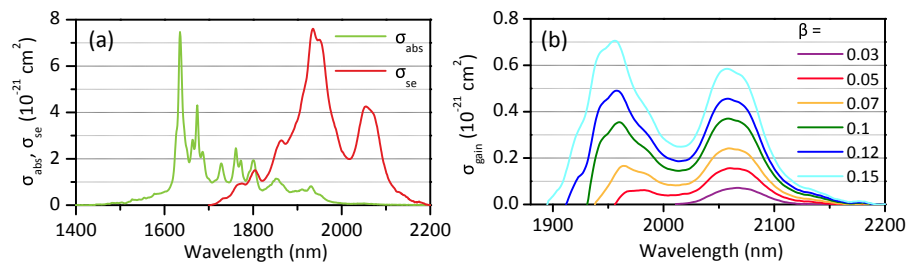


Fig. 1. (a) Absorption (σ_{abs}) and stimulated emission (σ_{se}) cross-sections of the Tm:Y₂O₃ ceramic for the ³H₆ ↔ ³F₄ transition, and (b) the corresponding gain cross-sections calculated by $\sigma_{\text{gain}} = \beta\sigma_{\text{se}} - (1 - \beta)\sigma_{\text{abs}}$ where β is the inversion ratio.

cross-section, σ_{se} , was thereafter calculated from the measured luminescence spectrum by using the Füchtbauer-Ladenburg equation [13]. As can be seen in Fig. 1(a), the maximum σ_{se} amounts to $7.6 \times 10^{-21} \text{ cm}^2$ at 1935 nm. The secondary peak at 2054 nm exhibits a $\sigma_{se} = 4.3 \times 10^{-21} \text{ cm}^2$. Based on the absorption and emission cross-sections, the gain cross-section was calculated using the equation: $\sigma_{gain} = \beta\sigma_{se} - (1 - \beta)\sigma_{abs}$, where $\beta = N(^3F_4)/N_{Tm}$ is the inversion ratio. As shown in Fig. 1(b), a smooth and broad gain profile extending from 1960 to 2150 nm is obtained, most pronounced for $\beta = 0.05$.

3. Laser setup and continuous-wave (CW) operation

An astigmatically compensated X-shaped cavity (see Fig. 2) was employed to study the CW and passively mode-locked Tm:Y₂O₃ ceramic laser. A 1627-nm Raman fiber laser with a beam quality (M^2 -factor) of 1.08 was employed for in-band pumping of the Tm³⁺ ions utilizing the main absorption peak of $^3H_6 \rightarrow ^3F_4$ transition, see Fig. 1(a). The corresponding absorption cross-section at 1627 nm amounted to $5 \times 10^{-21} \text{ cm}^2$. The collimated pump beam was focused into the ceramic using a spherical lens ($f = 75 \text{ mm}$), resulting in a beam radius of around $22 \mu\text{m}$. A 3-mm-thick 2 at.% Tm:Y₂O₃ ceramic sample with uncoated $3 \times 3 \text{ mm}^2$ aperture, was placed in the cavity at Brewster's angle. The sample, tightly mounted in a Cu holder, was water-cooled to $14 \text{ }^\circ\text{C}$ to mitigate the thermal load. CW and tunable laser operation was studied by using a simplified version of the cavity which comprised two dichroic folding mirrors (M1 and M2, with a radius of curvature of $R_{OC} = -100 \text{ mm}$), a flat rear reflector M4, and plane-wedged output couplers (OCs) with transmission of 1%, 1.5%, 3% and 5%. A 2-mm-thick quartz plate acting as a Lyot filter (LF) was inserted close to the OC and also at Brewster's angle. To study the mode-locking performance, a plano-concave mirror (M3) with $R_{OC} = -100 \text{ mm}$ was used to create a second beam waist of $\sim 60 \mu\text{m}$ radius on the saturable absorber (SA) which acted in this case as an end mirror. The SA used for starting and stabilizing the mode-locking, was a GaSb-based SESAM with a high linear reflectivity of $\sim 97\%$ at 2080 nm [14–16]. Flat chirped mirrors offering group delay dispersion (GDD) of -125 fs^2 (CM1 and CM2) and -875 fs^2 (CM3) per bounce at 2060 nm were employed for dispersion compensation. Using the ABCD formalism, the calculated laser beam radius on the Tm:Y₂O₃ ceramic was $35 \mu\text{m} \times 62 \mu\text{m}$ in the sagittal and tangential planes, respectively.

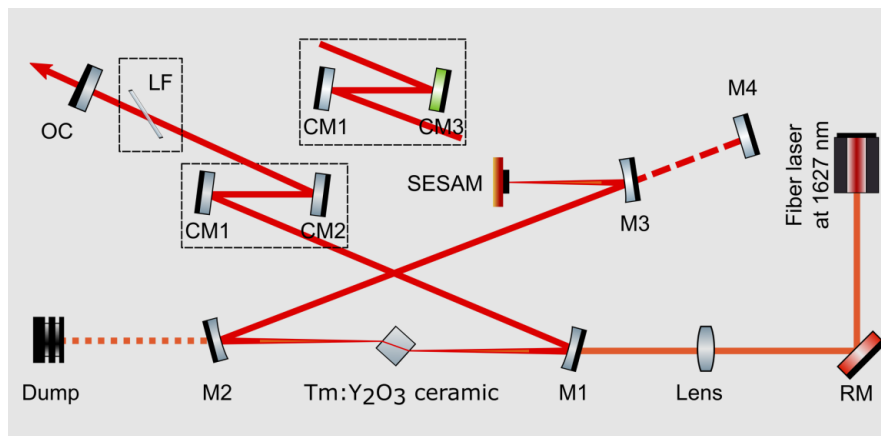


Fig. 2. Schematic of the CW and SESAM mode-locked Tm:Y₂O₃ ceramic laser in-band pumped by a 1627-nm Raman fiber laser. (RM: reflective mirrors; M1-M4: cavity mirrors; LF: Lyot filter; CM1-CM3: chirped mirrors; OC: output coupler).

At first, we studied the CW and wavelength-tunable laser performance of the Tm:Y₂O₃ ceramic laser with different OCs. The single-pass absorption under lasing conditions was measured to be 52%. As shown in Fig. 3(a), a maximum output power of 0.8 W was achieved with $T_{OC} = 5\%$ under an absorbed pump power of 2.04 W, corresponding to a laser slope efficiency of 44.7%. With decreasing the OC transmission, an obvious red shift of the wavelength was observed from 1954.4 to 2052.2 nm [see Fig. 3(b)], which is a typical feature of such a quasi-three-level Tm-laser system due to the enhanced reabsorption effect. The latter is related to a higher ground state population due to the recycling effect for the case of lower OC transmission [7].

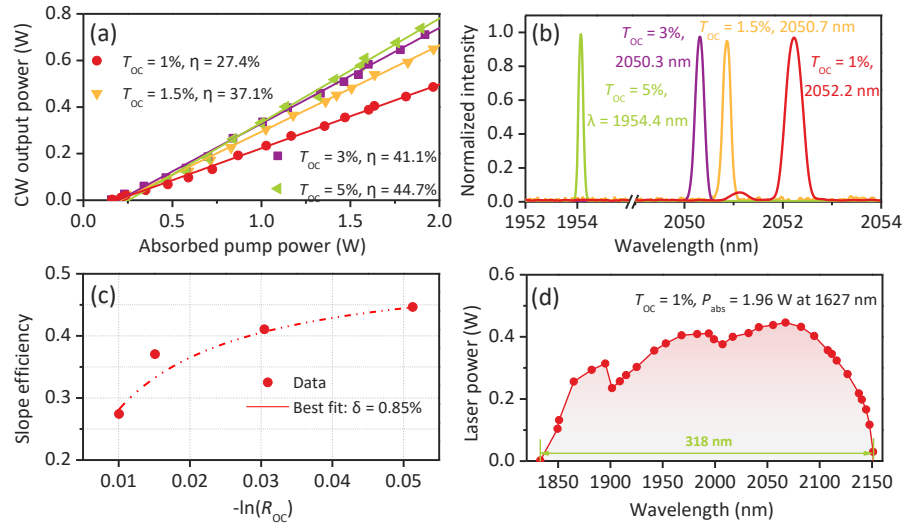


Fig. 3. CW and wavelength-tuning performance of the Tm:Y₂O₃ ceramic laser: (a) Output power versus absorbed pump power for different output coupler transmissions, η : the slope efficiency; (b) Optical spectra with different OCs; (c) Caird plot of the laser slope efficiency with respect to the OC reflectivity, i.e., $-\ln(R_{OC})$; and (d) wavelength tuning curve with $T_{OC} = 1\%$ at 1.96-W absorbed pump power.

The round-trip passive cavity loss δ was estimated by using the Caird analysis [17], as shown in Fig. 3(c). The best fit of the slope efficiency as a function of $-\ln(R_{OC})$ yielded a value of $\delta = 0.85\%$. Using the home-made Lyot filter, wavelength tunability of the Tm:Y₂O₃ ceramic laser was studied for $T_{OC} = 1\%$. The tuning range shown in Fig. 3(d) extends from 1833 to 2151 nm, corresponding to 318 nm, which is even broader than that the 296 nm obtained by combing Tm:Sc₂O₃ and Tm:Lu₂O₃ in one laser cavity [9]. Such a broad and smooth tuning range is a further indication of the potential of the Tm:Y₂O₃ ceramic for ultrashort pulse generation.

4. Mode-locking of the Tm:Y₂O₃ ceramic laser

Employing the SESAM, mode-locking of the Tm:Y₂O₃ ceramic laser was studied with the cavity shown in Fig. 2. A pair of chirped mirrors (CM1 and CM3) was inserted in the long cavity arm for dispersion compensation, giving a physical cavity length of ~ 1.7 m. Taking into account the group velocity dispersion (GVD) of Tm:Y₂O₃ calculated from its Sellmeier equation [18], the total round-trip GDD amounted to ~ -2100 fs² at 2.06 μ m [see Fig. 4(a)].

The highest average output power of 0.59 W at ~ 86 MHz pulse repetition rate was achieved with the 3% OC under an absorbed pump power of 2.05-W, corresponding to a laser efficiency of 29% and a pulse energy of 6.9 nJ. This corresponds to an average fluence of ~ 2 mJ/cm² on the SESAM. Mode-locking was self-starting and stable for hours. As can be seen from Fig. 4(a), the

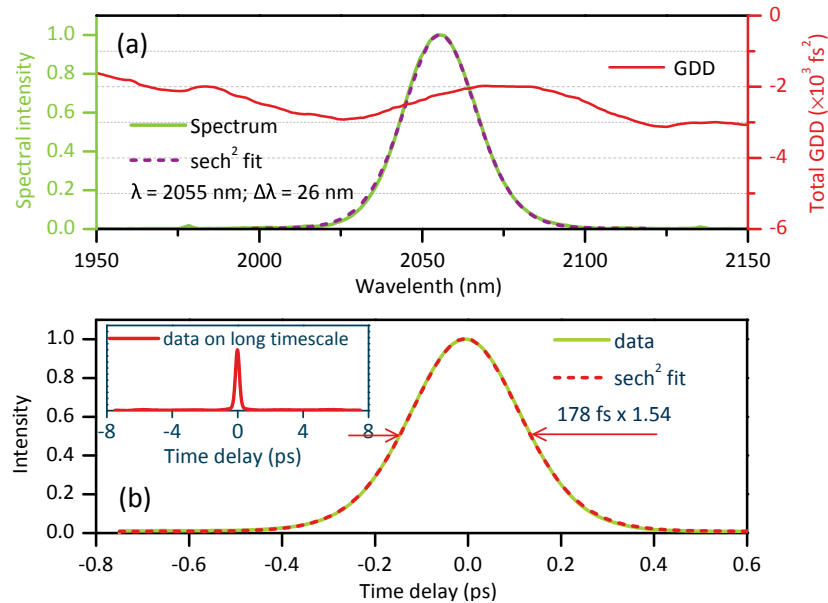


Fig. 4. (a) Optical spectrum and (b) autocorrelation trace of the SESAM mode-locked Tm:Y₂O₃ ceramic laser with $T_{OC} = 3\%$. The solid red curve in (a) represents the total round-trip GDD. *Inset* in (b): the corresponding long-scale (± 7.5 ps) autocorrelation trace.

optical spectrum centered at 2055 nm exhibits a perfect sech^2 -shaped profile with a full width at half maximum (FWHM) of 26 nm. The corresponding noncollinear autocorrelation trace is shown in Fig. 4(b). Fitting by a sech^2 -function for the temporal intensity profile, the obtained pulse duration amounted to 178 fs, giving a time-bandwidth product (TBP) of 0.33, close to the Fourier transform limit of 0.315. Single pulse operation of the mode-locked Tm:Y₂O₃ ceramic laser without any temporal satellites was confirmed by measuring a long time scale (± 7.5 ps) autocorrelation trace, see the inset of Fig. 4(b). In this case, the on-axis laser intensity on the Tm:Y₂O₃ ceramic was estimated to be 33 GW/cm². Pulse shortening should be still possible by further enhancing the self-phase modulation (SPM) effect with a lower OC transmission [19].

Thereafter, we employed the 1% OC in the mode-locked laser. In the soliton mode-locking regime, the pulse duration is proportional to the total round-trip GDD under the conditions of fixed SPM effect and pulse energy [19]. Thus the GDD was reduced by replacing CM3 with CM2. In this case, the total round-trip GDD dropped to ~ -650 fs² [see Fig. 5(a)]. The mode-locking was again self-starting, and an average output power of 0.26 W was achieved at an absorbed pump power of 2.11 W, yielding a pulse energy of ~ 3 nJ.

Figures 5(a) and 5(b) show the corresponding optical spectrum and interferometric autocorrelation trace, respectively. The optical spectrum centered at 2062 nm has a spectral FWHM of 60 nm. The peak-to-background ratio of the interferometric autocorrelation trace is exactly 8:1. By assuming a sech^2 -shaped envelope profile, the pulse duration amounted to 75 fs thus giving a TBP of 0.317, which is an indication of almost chirp-free pulses. Similarly, the clean pulse profile on a long time scale [see the inset of Fig. 5(b)] indicates steady-state mode-locking with single pulse operation. In comparison, the on-axis laser intensity on the ceramic has increased up to ~ 100 GW/cm².

To further characterize the stability of the mode-locked Tm:Y₂O₃ ceramic laser, we recorded the real-time pulse trains using a fast (> 10 GHz) photodiode and a digital oscilloscope. As shown in Fig. 5(c), the uniform pulse train recorded on a nanosecond time scale shows a good

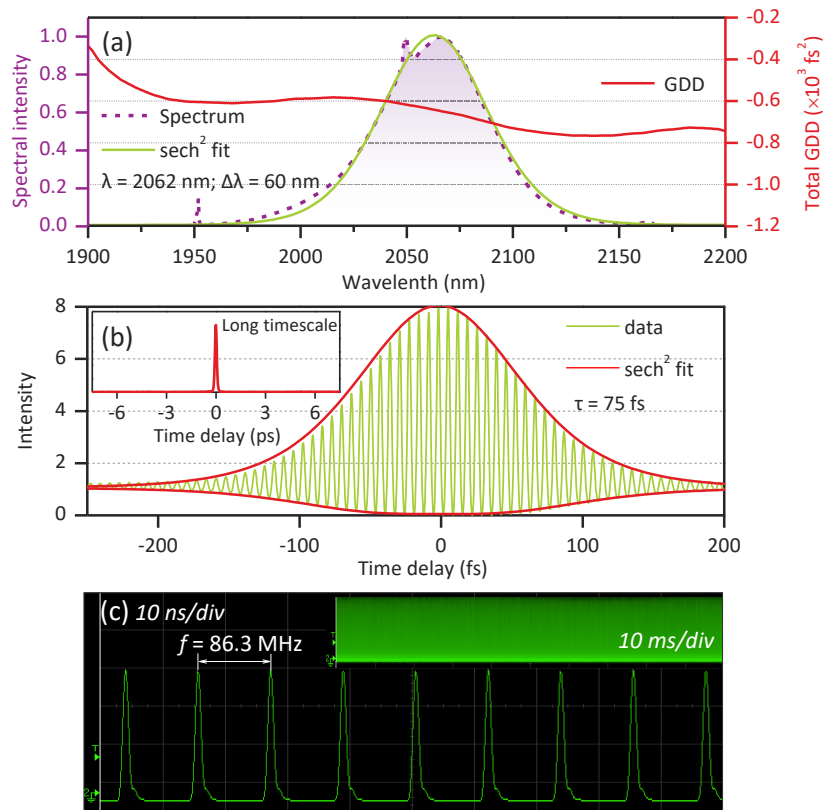


Fig. 5. (a) Optical spectrum, (b) interferometric autocorrelation trace, and (c) typical pulse train on a nanosecond time scale of the shortest pulses produced with $T_{OC} = 1\%$. The red line in (a) represents the calculated total round-trip GDD. *Insets* in (b) and (c): noncollinear autocorrelation trace on a long time scale of ± 7.5 ps, and pulse train on a millisecond time scale, respectively.

pulse-to-pulse stability. In addition, no Q-switching and multi-pulse instabilities were observed from the long-scale (10 ms/div) pulse train.

5. Conclusion

In conclusion, polycrystalline Y_2O_3 transparent ceramic doped with 2 at.% Tm^{3+} was fabricated by using vacuum sintering followed by hot isostatic pressing in an Ar atmosphere. The smooth and flat gain spectrum was instrumental for achieving unprecedented wavelength tunability. By in-band pumping with a Raman fiber laser at 1627 nm, the $Tm:Y_2O_3$ laser was tunable across 318 nm in the 2- μm spectral range using a Lyot filter. By employing a SESAM, the first femtosecond mode-locked $Tm:Y_2O_3$ laser has been realized, delivering sub-80-fs pulses at a central wavelength of 2062 nm with a spectral bandwidth of 60 nm. The average output power reached 0.59 W at longer pulse durations. High laser efficiency was achieved thanks to the in-band pumping. Further power scaling is feasible just by increasing the pump power since no roll-over was observed in the input-output characteristics in the CW regime. On the other hand, further pulse shortening might be still possible by optimizing the dispersion management and enhancing the SPM effects through reducing the OC transmission. Nevertheless, the broad spectral tuning range and the sub-80-fs pulses obtained in the present work indicate that polycrystalline sesquioxide transparent

ceramics doped with Tm^{3+} - or $\text{Tm}^{3+}, \text{Ho}^{3+}$ are promising candidates for development of power scalable femtosecond mode-locked bulk lasers in the 2- μm spectral range. Such sources will find various applications such as synchronous pumping of optical parametric oscillators (SPOPOs) to produce mid-IR wavelengths [20], generation of mid-IR frequency combs for spectroscopy in the molecular fingerprint region [21], seeding of Ho-doped laser amplifiers to produce high power/energy femtosecond pulses near 2 μm [22], and processing of transparent materials [23].

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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