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Sub-10 optical-cycle passively mode-locked Tm:$(Lu_{2/3}Sc_{1/3})_2O_3$ ceramic laser at 2 μm

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Abstract: A Tm-doped mixed sesquioxide ceramic laser is mode-locked near 2 μm using InGaAsSb quantum-well semiconductor saturable absorber and chirped mirrors for dispersion compensation. Maximum average output power of 175 mW is achieved for a pulse duration of 230 fs at a repetition rate of 78.9 MHz with a 3% output coupler. Applying a 0.2% output coupler pulses as short as 63 fs are generated at 2.057 μm.

OCIS codes: (140.4050) Mode-locked lasers; (140.3070) Infrared and far-infrared lasers; (160.3380) Laser materials.

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1. Introduction

Thulium (Tm^{3+}) based solid-state lasers generating ultrashort pulses in the eye-safe spectral region around 2 μm are currently actively investigated for potential applications in environmental monitoring [1] and material processing [2]. They have been already widely applied for frequency down-conversion into the mid-IR, as pump sources for synchronously pumped optical parametric oscillators [3]. As seeding sources for high power ultrafast amplifiers they are expected to play an important role also in high-harmonic and soft X-ray generation [4]. Bulk materials exhibit certain advantages over fibers [5] and few families of Tm^{3+}-doped laser gain media have been successfully employed so far in passively mode-locked bulk solid-state lasers emitting in the 2-μm spectral range. The monoclinic tungstates Tm:$KLu(WO_4)_2$ (KLuW) and Tm:$MgWO_4$ (MgW), exhibiting large emission cross-sections and bandwidths, have been one of the most successful ones generating pulses as short as 141 fs [6] and 86 fs [5]. However, in the quasi-3-level Tm-laser system what counts are the gain cross-sections and bandwidths. Another family known for their superior thermo-mechanical and thermo-optical properties are the cubic (C-type, bixbyite structure) rare-earth sesquioxides A_2O_3 (where A = Lu, Y, Sc). Employing a Tm:Sc_2O_3 crystal, 218 fs pulses were generated with an ion-implanted InGaAsSb-quantum-well based semiconductor saturable absorber mirror (SESAM) [7] and 166 fs by Kerr-lens mode-locking [8]. With Single-Walled Carbon Nanotube Saturable Absorbers (SWCNT-SAs) pulses as short as 175 fs were generated employing a Tm:Lu_2O_3 crystal [9]. Pulse durations as short as 105 fs were mentioned in a short report [10] employing the mixed $LuScO_3$ crystal with the same SESAM as in [7], however, the optical spectrum exhibited a continuous-wave (CW) component.

Due to the high melting point of the cubic A_2O_3 crystals (e.g. 2450°C for Lu_2O_3), their growth typically requires the use of expensive rhenium (Rh) crucibles and Rh could be the source of the observed crystal coloration [11]. The growth of large-volume highly Tm^{3+}-doped single crystals with high optical quality is complicated and the fabrication of transparent sesquioxide ceramics is a promising alternative. There are few reports on CW lasing of transparent Tm:Lu_2O_3 ceramics in different schemes including thin-disk lasers [12–14]. Femtosecond lasers based on Tm:Lu_2O_3 ceramics have also been investigated: using

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single-layer graphene for mode-locking resulted in pulse duration of ~410 fs [15] while SESAM mode-locking produced pulse durations as short as 180 fs [16].

Recently, we reported on fabrication, spectroscopy and CW laser operation of novel Tm^{3+}:$(Lu_{2/3}Sc_{1/3})_2O_3$ (shortly Tm:LuScO) mixed ceramics [17]. As a consequence of the compositional disorder (inhomogeneous spectral line broadening), mixed Tm:LuScO ceramics exhibit very broad absorption and emission bands [17] similar to the mixed single crystals [18]. Moreover, pumped by a laser diode at 802 nm, a Watt level Tm:LuScO ceramic micro-laser at 2.1 μm was demonstrated with a slope efficiency of 24% [17]. These promising spectroscopic features and laser performance motivated us to investigate passive mode-locking of such Tm:LuScO ceramics. Here we report on the realization of this regime employing a SESAM and chirped mirrors (CMs) for dispersion management, setting a pulse duration record for a mode-locked bulk solid-state laser operating in the 2-μm spectral range.

2. Tm:LuScO mixed ceramics

High quality Tm:$(Lu,Sc)_2O_3$ ceramics were fabricated by the Hot Isostatic Pressing (HIP) sintering method using powders of Sc_2O_3, Lu_2O_3, and Tm_2O_3 (purity: 99.99%, Alfa Aesar). The raw materials, with a stoichiometric amount of 100 at. % Lu + Sc (taken in a proportion of Lu:Sc = 2:1, the only ratio for which good optical quality could be obtained) and 5 at. % Tm over it, were mixed uniformly by ball milling for 24 h, dried for 6 h at 70°C, sieved, dry-pressed at 10 MPa, and cold isostatically pressed at 200 MPa. The green bodies of Tm:$(Lu,Sc)_2O_3$ ceramics were first pre-sintered at 1750°C for 10 h under vacuum (pressure, $P < 10^{-3}$ Pa) to densify the preforms. For further densification, the pre-sintered ceramic samples were post-sintered by HIP at 1800°C for 2 h in an Ar atmosphere (P = 195 MPa) to eliminate the closed pores around the grain boundaries. Finally, the ceramics were annealed at 1500°C for 10 h in an O_2 atmosphere to eliminate the oxygen vacancies and remove internal stresses. The composition of the ceramics can be represented as 4.76 at. % Tm:$(Lu_{2/3}Sc_{1/3})_2O_3$ or shortly Tm:LuScO. Samples with a diameter of 15 mm and a thickness of 5 mm were obtained, see Fig. 1(a). For the laser experiments, a 2.95 mm thick active element was cut and its input and output surfaces (aperture ~3 × 3 mm^2) were polished to laser-grade quality.

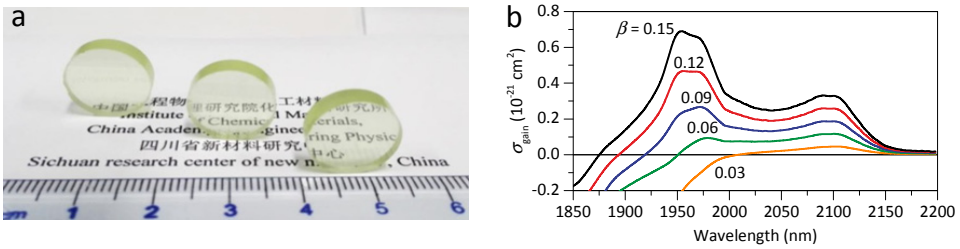

Fig. 1. (a) The fabricated 4.76 at.% Tm:LuScO mixed ceramic disks (laser-grade-polished) and (b) calculated gain cross section σ_{gain} for different inversion levels β of the Tm:LuScO mixed ceramic in the 2-μm spectral range.

In order to estimate the potential spectral emission of the Tm:LuScO ceramics, the gain cross section $\sigma_{gain} = \beta\sigma_e - (1-\beta)\sigma_a$ where σ_e and σ_a denote the emission and absorption cross sections, respectively, was calculated for several values of the population inversion parameter β, see Fig. 1(b) [17]. β is the ratio of the excited Tm^{3+}-ions in the $^3F_4$ manifold to the total Tm^{3+}-ion density. All the Tm:LuScO ceramics cross sections are very similar to those of their single crystal counterparts [18]. Similar to the mixed crystals, the mixed ceramics exhibit smoother wavelength dependence compared with the single ceramics which is potentially advantageous for enhancing the mode-locked pulse bandwidth. For low inversion levels emission of the free running laser is expected above 2 μm. This is beneficial for femtosecond pulse generation because the strong water absorption below 1.95 μm is avoided.

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3. Experimental setup

The experimental setup of the Tm:LuScO ceramic laser shown in Fig. 2 was based on an X-shaped cavity. A narrow-linewidth Ti:sapphire laser tuned to 0.795 µm was used for pumping in the absorption peak corresponding to the $^3H_6 \rightarrow {}^3H_4$ transition. The single pass absorption amounted to ~74%. Note that this absorption band at 0.74-0.83 µm is rather broad (FWHM = 25 nm) and thus suitable for AlGaAs laser diode pumping [17]. The pump beam was focused with a 70 mm lens. The Tm:LuScO ceramic sample was placed at Brewster angle between two dichroic folding mirrors M_1 and M_2 with a separation of ~100 mm. It was wrapped in In-foil for good contact (4 lateral sides) with the water cooled (14°C) Cu-holder.

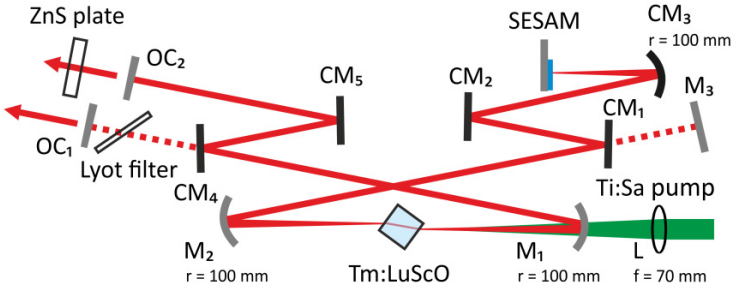

Fig. 2. Scheme of the Tm:LuScO mixed ceramic laser (L: lens, M: dichroic mirror, CM: chirped mirror, OC: output coupler, r: radius of curvature, f: focal length).

For CW tunable operation, a 3.2 mm thick quartz plate was inserted as a Lyot filter close to the wedged output coupler OC_1 (transmission $T = 1.5\%$). A plain mirror M_3 was used as an end mirror in this case leading to a cavity length of ~150 cm. In the mode-locking experiments, 5 chirped mirrors ($CM_{1\text{-}5}$) with a group-delay dispersion of GDD = –125 fs^2 per bounce were used for intracavity dispersion management. This leads to a total GDD of –1250 fs^2 per round-trip. An InGaAsSb quantum-well based SESAM with 2 quantum-wells and 50 nm thick cap layer design [19] served as an end mirror in this case leading to a cavity length of 190 cm. Output couplers OC_2 with $T = 3.0\%$, 1.5%, 0.5% and 0.2% were tested.

4. Experimental results and discussion

Smooth and continuous tuning from 1.978 to 2.108 µm was achieved with the 1.5% OC in the CW regime as shown in Fig. 3 for an absorbed power of P_{abs} = 1.09 W. The total CW tuning range of 130 nm is limited on the long-wave side by the mirror transmissions (3% for the OC).

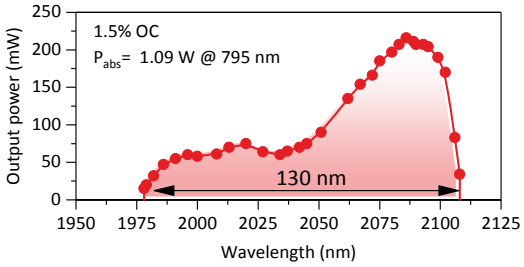

Fig. 3. Tuning of the CW Tm:LuScO mixed ceramic laser with a Lyot filter and 1.5% OC.

Mode-locking with the SESAM was self-starting and stable for hours without disruption during the daily operation with all the OCs. The performance applying different OCs was compared at a fixed incident power of 1.67 W (measured in front of L) corresponding to P_{abs}

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~1.2 W. The non-collinear autocorrelation traces and the optical spectra of the mode-locked laser are shown in Fig. 4 and all relevant parameters are summarized in Table 1. The highest average output power of 175 mW was achieved with the 3.0% OC for a pulse duration of τ = 230 fs (FWHM for a sech^2-pulse shape), Fig. 4(a). The OCs with decreasing transmission enabled substantial pulse shortening albeit at the expense of the average output power.

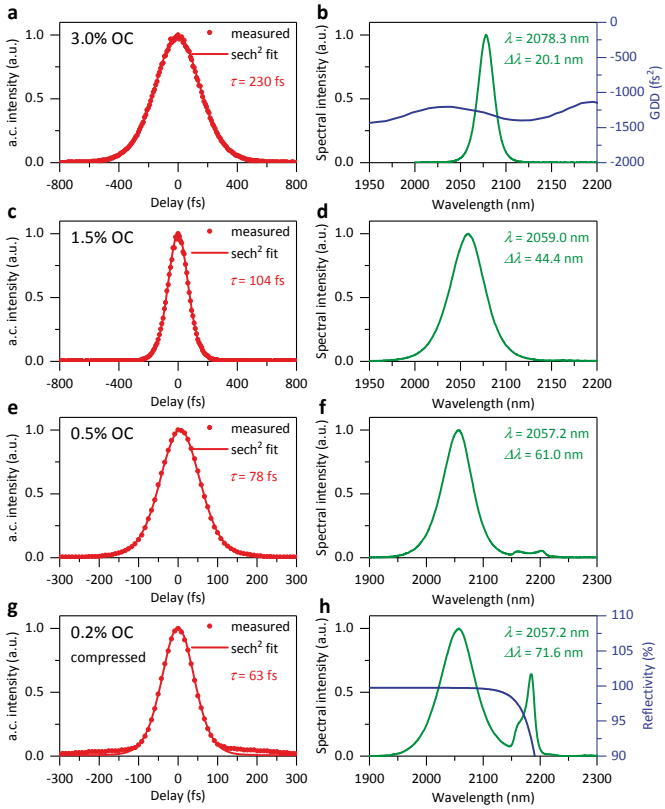

Fig. 4. Autocorrelation traces (a, c, e and g) measured by type-I second-harmonic generation in a 3-mm thick β-BaB_2O_4 crystal and optical spectra (b, d, f and h) of the mode-locked Tm:LuScO mixed ceramic laser measured with a 0.5 nm resolution rotating grating spectrometer for different OCs. Blue lines indicate calculated round trip GDD of the chirped mirrors and the 0.2% OC reflectivity, in (b) and (h), respectively.

Table 1. Mode-locking results with the Tm:LuScO ceramics laser with different OCs (P_{out}, average output power; τ, pulse duration; $\Delta\lambda$, spectra bandwidth as FWHM; TBP, time bandwidth product).

OC	P_{out} [mW]	τ [fs]	$\Delta\lambda$ [nm]	TBP
3.0%	175	230	20.1	0.321
1.5%	120	104	44.4	0.327
0.5%	50	78	61.0	0.337
0.2%	34	74	71.6	0.375
0.2% (compressed)		63	71.6	0.320

The shortest pulses with a FWHM of τ = 74 fs and ultimate stability were achieved using the 0.2% OC, at an average output power of 34 mW. No post- or pre-pulses were observed which was confirmed by a measurement in a longer (50 ps) time window. The spectrum was 71.6 nm broad and centered at ~2.057 μm as shown in Fig. 4(h). The secondary peak at longer wavelengths is considered to be leakage through the OC whose transmission increases

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to about 10% at 2.184 µm. The resulting time-bandwidth product (TBP) was 0.375, i.e. higher compared with the other OCs which yielded nearly transform-limited pulses. Thus a 3 mm thick ZnS plate (GDD = 462 fs^2) was placed after OC_2 as an extra-cavity compression element. The pulse could be compressed in this way to 63 fs which corresponds to ~9.2 optical cycles. It is also important to point out that the SESAM applied in the experiment has a rather long relaxation time of ~20 ps [19]. Only picosecond or sub-picosecond pulses down to ~650 fs could be generated previously with such a SESAM [20,21]. This indicates that the Kerr effect plays an important role for the sub-100 fs operation in the present work. However, pure Kerr-lens mode-locking was not achieved with an end mirror instead of the SESAM.

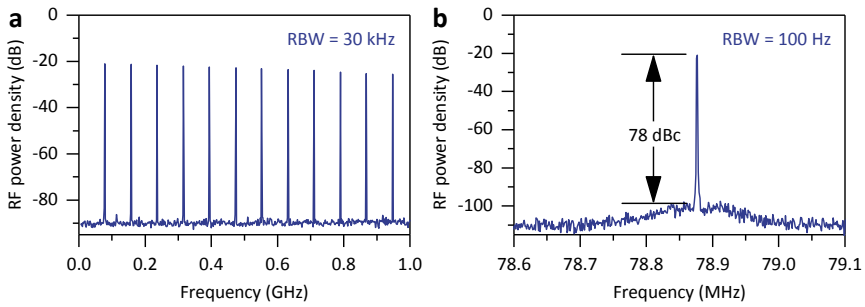

Fig. 5. Radio frequency spectra of the SESAM mode-locked Tm:LuScO ceramic laser with the 0.2% OC: (a) 1-GHz-wide span, (b) fundamental beat note (RBW: resolution bandwidth).

To further characterize the stability of the mode-locked Tm:LuScO ceramic laser, the radio frequency (RF) spectra were measured. With different OCs the performance was similar and the results with 0.2% OC are shown in Fig. 5. The wide-span RF measurement with resolution bandwidth of 30 kHz in Fig. 5(a) shows almost constant harmonics intensity within the 1 GHz span range. The narrow-band fundamental beat note was at 78.9 MHz with a high extinction ratio of 78 dB above noise level as shown in Fig. 5(b), measured with a resolution bandwidth of 100 Hz. Both RF spectra indicate clean CW mode-locking without Q-switching instabilities or any multi-pulse behavior [22].

5. Conclusion

In conclusion, sub-10 optical-cycle pulses were generated for the first time with a mode-locked 2 µm bulk solid-state laser. A Tm:LuScO (4.76 at.% Tm:$(Lu_{2/3}Sc_{1/3})_2O_3$) mixed ceramic laser passively mode-locked by a SESAM produced nearly transform-limited pulses as short as 63 fs at ~2.057 µm after careful optimization of the intracavity dispersion using chirped mirrors and minor extracavity compression in a lossless bulk material. The substantial pulse shortening achieved in comparison to previous work with sesquioxide crystalline or ceramic hosts is attributed to the flat and smooth gain spectra of Tm:LuScO mixed ceramics.

Further possibilities for pulse shortening are seen in the optimization of the Lu_2O_3 and Sc_2O_3 mixing ratio and/or co-doping with Holmium for further spectral gain broadening. In such a way, we believe that there is still great potential for such mixed sesquioxide ceramic systems to generate sub-50 fs or even few-cycle pulses around 2 µm.

Acknowledgments

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