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Sub-6 optical-cycle Kerr-lens mode-locked Tm:Lu_2O_3 and Tm:Sc_2O_3 combined gain media laser at 2.1 μm

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Abstract: We present a combined gain media Kerr-lens mode-locked laser based on a Tm:Lu_2O_3 ceramic and a Tm:Sc_2O_3 single crystal. Pulses as short as 41 fs, corresponding to less than 6 optical cycles, were obtained with an average output power of 42 mW at a wavelength of 2.1 μm and a repetition rate of 93.3 MHz. Furthermore, a maximum average power of 316 mW with a pulse duration of 73 fs was achieved.

1. Introduction

Tm-doped materials are excellent candidates as gain media for high power 2 μm lasers. This is due to their highly efficient "two-for-one" pumping scheme enabled by cross relaxation between adjacent Tm^{3+}-ions when pumped by commercially available high power laser diodes around 790 nm. During the last decade, the rise of novel Tm-doped materials with broad gain bandwidth enabled the development of Tm-doped lasers with sub-100 fs pulse duration. Pulses as short as 84 fs and 78 fs were generated from a graphene mode-locked Tm:CNNGG laser at 2018 nm [1] and a single-walled carbon nanotube (SWCNT) mode-locked Tm:CLNGG laser at 2017 nm [2], respectively. Even shorter pulses of 76-fs duration were achieved using SWCNT assisted Kerr-lens mode-locked Tm:$MgWO_4$ laser at 2037 nm [3]. Also Tm-doped cubic sesquioxide (Tm:RE_2O_3, RE = Lu, Y or Sc) materials are attractive for high power ultrashort pulse lasers. This is due to their superior spectroscopic, thermo-mechanical and thermo-optical properties [4,5]. They show high thermal conductivity and low thermo-optic coefficients. Moreover, the gain band of Tm-doped sesquioxides is centered at extremely long wavelengths, which avoids problems with strong water vapor absorption around 2 μm and also reduces reabsorption. The combination of both effects facilitates very broad effective gain bandwidths in Tm-doped sesquioxides. Employing a Tm:Lu_2O_3 single crystal, pulses as short as 175 fs were generated from a SWCNT mode-locked laser at 2070 nm [6] and the use of a Tm:Sc_2O_3 single crystal enabled pulses as short as 218 fs from a semiconductor saturable absorber mirror (SESAM) mode-locked laser at 2113 nm [7]. We also achieved pulses as short as 72-fs from a Kerr-lens mode-locked (KLM) Tm:Sc_2O_3 single crystal laser at 2108 nm [8]. More recently, with the appearance of mixed sesquioxide materials, even shorter pulses were reported. Pulses as short as 63 fs and 54 fs were generated from a SESAM mode-locked Tm:$LuScO_3$ ceramic laser at 2057 nm [9] and a SESAM mode-locked Tm:$LuYO_3$ ceramic laser at 2040 nm [10], respectively. These mixed materials show broad and smooth gain spectra due to their compositional disorder. However, they exhibit reduced cross sections and thermal conductivities, which imposes difficulties for high power laser operation. In this study, we present a combined gain media KLM laser [11,12] based on the simultaneous use of a Tm:Lu_2O_3 ceramic and a Tm:Sc_2O_3 single crystal in the same cavity. This combined gain media laser enables a broad and flat gain spectrum extending from

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1850 nm to 2200 nm without affecting the thermal conductivity of the gain material. With this approach, we achieved pulse durations as short as 41 fs, i.e. sub-6 optical cycles, after external compression.

2. Combined gain media laser

The idea of a combined gain media laser is very simple: different gain materials are used simultaneously in the same cavity, so that the laser can benefit from the gain spectra of both materials. Figure 1 shows emission and absorption cross sections of Tm:Lu_2O_3 and Tm:Sc_2O_3. Their emission cross sections are located in the range between 1850 nm and 2200 nm, but due to the stronger crystal field strength on the smaller Sc^{3+} site as compared to the larger Lu^{3+}, the emission peaks of Tm:Sc_2O_3 are red shifted by roughly 30 nm compared to those of Tm:Lu_2O_3. The effective gain cross section σ_{eff} resulting from the use of Tm:Lu_2O_3 and Tm:Sc_2O_3 as combined gain media can be described as below [11],

$$\boldsymbol{\sigma_{eff}}(\omega) = \boldsymbol{\alpha\beta_1\sigma_{e1}} - \alpha(\mathbf{1} - \boldsymbol{\beta}_1)\boldsymbol{\sigma_{1a}} + (\mathbf{1} - \alpha)\boldsymbol{\beta_2\sigma_{2e}} - (\mathbf{1} - \alpha)(\mathbf{1} - \boldsymbol{\beta}_2)\boldsymbol{\sigma_{2a}}$$

where σ_{e1}, σ_{e2}, σ_{a1} and σ_{a2} are emission cross sections and absorption cross sections of Tm:Lu_2O_3 and Tm:Sc_2O_3, respectively. β_1 and β_2 indicate the population inversion ratios of the two gain media. α indicates the ratio of the Tm^{3+}-ions in the Tm:Lu_2O_3 gain medium against the total amount of Tm^{3+}-ions in both gain media. The effective gain shape strongly depends on the ratios α and β. While α can be tuned by optimizing the doping concentrations and/or the thicknesses of both gain media, the inversion parameter β is mainly affected by the pump intensity. By a proper choice of all parameters, thus an increased gain bandwidth with a flat and smooth shape can be obtained.

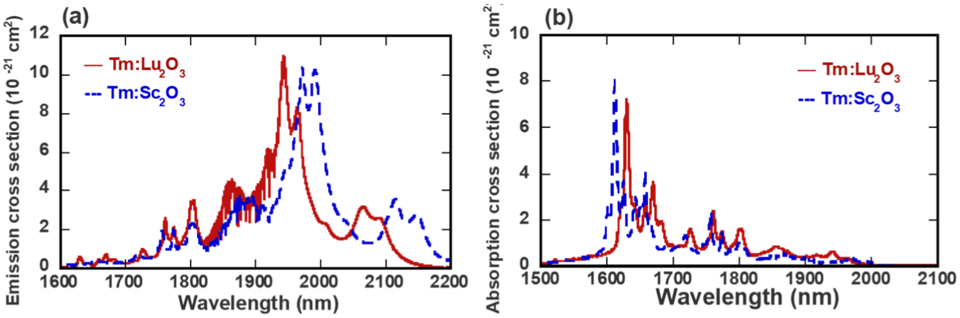

Fig. 1. (a) Emission cross sections of Tm:Lu_2O_3 (red solid) and Tm:Sc_2O_3 (blue dashed), (b) Absorption cross sections of Tm:Lu_2O_3 (red solid) and Tm:Sc_2O_3 (blue dashed)

3. Experimental setup

Figure 2(a) shows the experimental set up of our combined gain media KLM laser. We used an astigmatism-compensated Z-shaped cavity composed by two curved folding mirrors (R = 100 mm, HR>99.9% at 1850-2200 nm), different chirped mirrors with varying negative GDD values for dispersion control and a wedged output coupler (OC). A 4 mm thick Tm(4 at.%):Lu_2O_3 ceramic (n=1.90) and a 3.7 mm thick Tm(1 at.%):Sc_2O_3 single crystal (n=1.94) without AR coating served as the gain media. They were in physical contact to each other, mounted in a Peltier cooled copper heatsink and placed in the focus between the two curved folding mirrors at Brewster's angle. We used a home-built 1611 nm Er:Yb-doped all fiber MOPA [13] as a pump source. At the pumping wavelength, the Tm:Lu_2O_3 has significantly lower absorption cross sections than the Tm:Sc_2O_3 (cf. Figure 1(b)), which was balanced to a large extend by the higher doping concentration of the Tm:Lu_2O_3 ceramic. Thus, the estimated small signal absorptions were 58% and 68% for the Tm:Lu_2O_3 and the Tm:Sc_2O_3, respectively. In order to further balance pump absorption, the Tm:Lu_2O_3 was placed at side facing the pump source and the Tm:Sc_2O_3 at the opposite side, so that the Tm:Sc_2O_3 was pumped by the residual pump beam passing the Tm:Lu_2O_3. The estimated diameters of the pump laser mode and the cavity fundamental mode at the focusing point were 43 × 43 μm^2 and 66 × 65 μm^2, respectively.

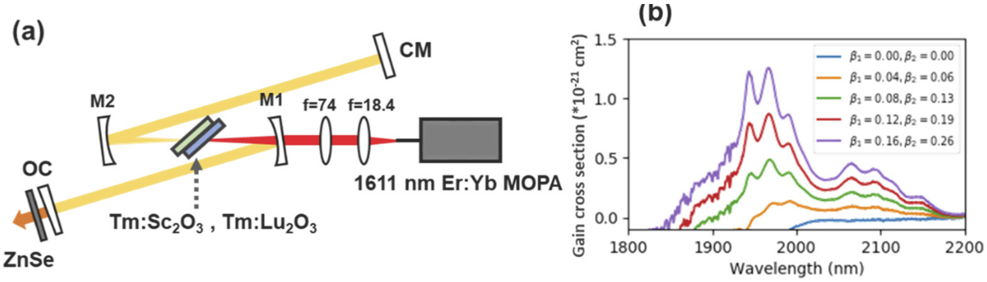

Fig. 2. (a) Experimental set up of the combined gain media KLM laser. (b) Calculated effective gain cross sections assuming different inversion population ratios β_1 and β_2. Note that the ratio between β_1 and β_2 must remain constant as both gain media are pumped by the same pump source.

The effective gain cross sections under these conditions are shown in Fig. 2(b) and show a broad and smooth gain band reaching from 1950nm to 2160 nm. Mode-locked operation was achieved by soft-aperture Kerr-lens mode locking. The output pulses were externally dispersion compensated by passing a 3 mm ZnSe window.

4. Results and discussion

At first, we demonstrated CW and wavelength tunable operation. The output power as a function of absorbed pump power and the free running spectra with different OCs are shown in Figs. 3(a) and 3(b). We found an increasing slope efficiency with increasing OC transmittance in the range of the available mirrors transmittances. The highest slope efficiency of 35.1% was obtained for the 9% OC. In a second experiment, we demonstrated wavelength tunable laser operation using an IR grade fused silica prism as a wavelength tuning element. We obtained an extremely broad tuning range of 296 nm from 1874nm to 2171 nm as shown in Fig. 3(c) which should support very short pulses in the forthcoming mode locking experiments.

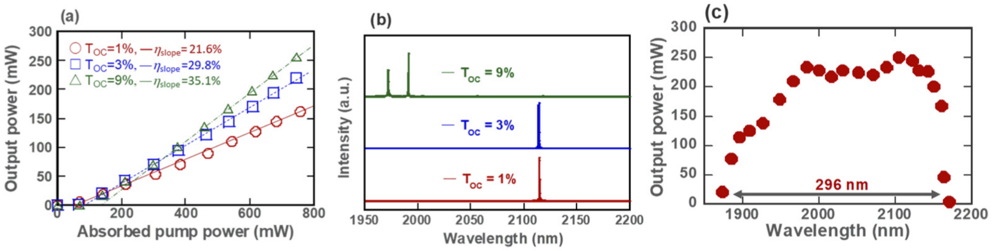

Fig. 3. (a) Output power as a function of pump power with different OCs, (b) free running spectra for different OCs, (c) wavelength tunability of a combined gain media laser with 1% OC under 1.7 W pumping.

Next, we performed KLM experiments using different OCs of 3%, 1% and 0.5% transmittance. KLM operation was initiated by moving mirror M2 (see Fig. 2(a)). We estimated the GDD at

2.1 μm resulting from the gain media to be ~ -160 fs^2 and ~-420 fs^2 in the Tm:Lu$_2$O$_3$ ceramic and the Tm:Sc$_2$O$_3$ crystal, respectively.

Using an OC with 3% transmittance and a total cavity GDD of ~-1580fs^2 obtained by using a chirped mirror (HR>99.9% at 1950-2200 nm) with a GDD ~-1000 fs^2, we obtained a maximum average output power of 316 mW at a pump power of 2 W. Under these conditions, the pulse duration after compression was 73 fs determined by SHG intensity autocorrelation as shown in Fig. 4(a) and the repetition rate amounted 93.2 MHz. The corresponding pulse energy and peak power reached 3.4 nJ and 46.4 kW, respectively. The optical spectrum of the mode-locked laser is shown in Fig. 4(b) and exhibits a spectral bandwidth (FWHM) of 64 nm around a center wavelength of 2090nm, implying a time bandwidth product of 0.321 close to the transform limit for sech^2-pulses of 0.315.

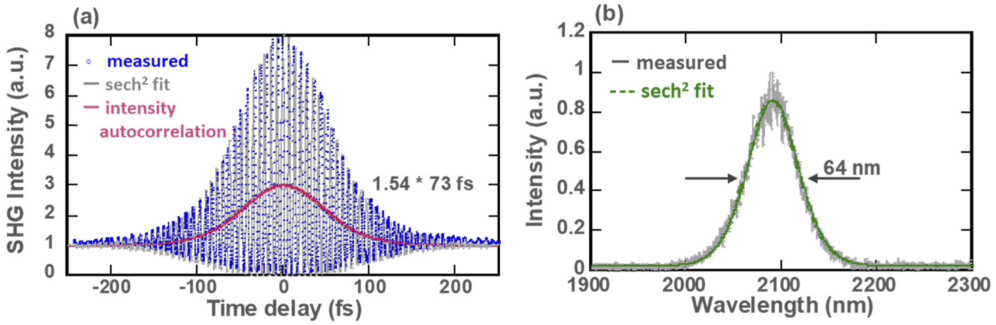

Fig. 4. (a) SHG intensity autocorrelation trace after compression and (b) optical spectrum using the 3% OC

By reducing the output coupler transmission and the cavity dispersion, we aimed to further reduce the pulse duration. Using an 1% OC and a chirped mirror with a lower GDD of ~-300 fs^2 (HR>99.9% at 1950-2450 nm) and a pump power of 1.7 W, we obtained an average output power of 81 mW. In this case, the pulse duration directly after the cavity was reduced to 67 fs at an unchanged repetition rate of 93.2 MHz. After external compression, pulses as short as 52 fs were obtained as evidenced by the autocorrelation trace shown in Fig. 5(a). The corresponding pulse energy and peak power were 0.87 nJ and 16.7 kW, respectively. The spectral bandwidth was 88.8 nm centered around 2095nm (Fig. 5(b)). The broadened spectrum from 2000nm to 2200 nm beyond the gain bandwidth was obtained by the combination of the effect of large self-phase modulation and the spectra of the combined gain media. The time bandwidth product was 0.315, indicating transform limited pulses.

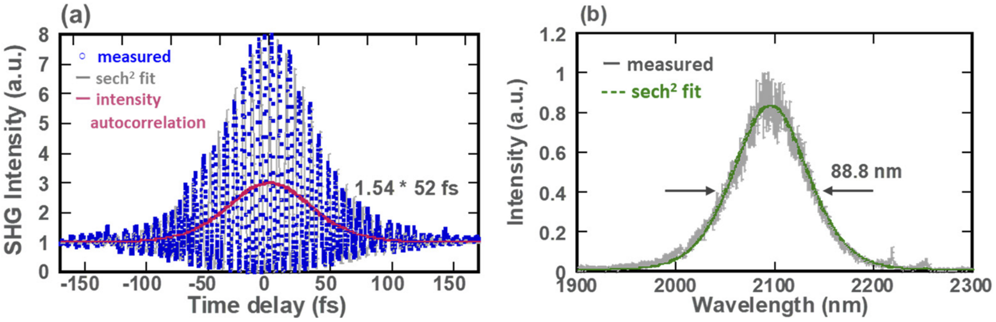

Fig. 5. (a) SHG intensity autocorrelation trace after compression and (b) optical spectrum using the 1% OC

Finally, in order to obtain the shortest pulses at the expense of a reduced output power, we increased the quality-factor (Q-factor) of the cavity by replacing the 1% OC with a 0.5% OC. In this configuration, we achieved the shortest pulse duration of 41 fs after compression with an average output power of 42 mW at a pump power of 1.9 W. The corresponding autocorrelation trace is shown in Fig. 6(a). Figure 6(b) shows the radio frequency (RF) spectrum with a high signal to noise ratio of more than 70 dB at the fundamental repetition rate of 93.3 MHz. Pulse energy and peak power amounted to 0.45 nJ and 11.0 kW, respectively. The spectral bandwidth (FWHM) was 104.5 nm with a center wavelength of 2094nm. In the optical spectrum shown in Fig. 6(c), additional spectral components are found in the wavelength range between 2210 nm and 2350 nm where both gain media show no gain. We attribute this to Raman assisted spectral broadening [14] caused by an increased intracavity peak power resulting from the high Q-factor of the cavity, as it was only observed for the 0.5% OC. In fact, despite a reduced average power, the laser using the 0.5% OC shows the highest intracavity peak power of 2.20 MW compared to 1.55 MW and 1.70 MW at 3% and 1% OC, respectively. The structural characteristics of the optical spectrum of the mode-locked pulses as well as the cavity conditions are similar to previous reports of spectral broadening due to intra-pulse SRS [8,14]. The transform limited pulse duration calculated using only the sech^2-shaped soliton mode-locked component of the optical spectrum is 45 fs. In contrast, using the whole spectrum, the transform limited pulse duration reaches ~29 fs as shown in Fig. 6(d). We measured a pulse width of 41 fs, which indicates that the additional spectral component contributes somewhat to pulse shortening. It should, however, be noted, that the additional spectral components act as a nonlinear loss for the mode-locked pulses, as they do not contribute to stimulated emission due to the lack of

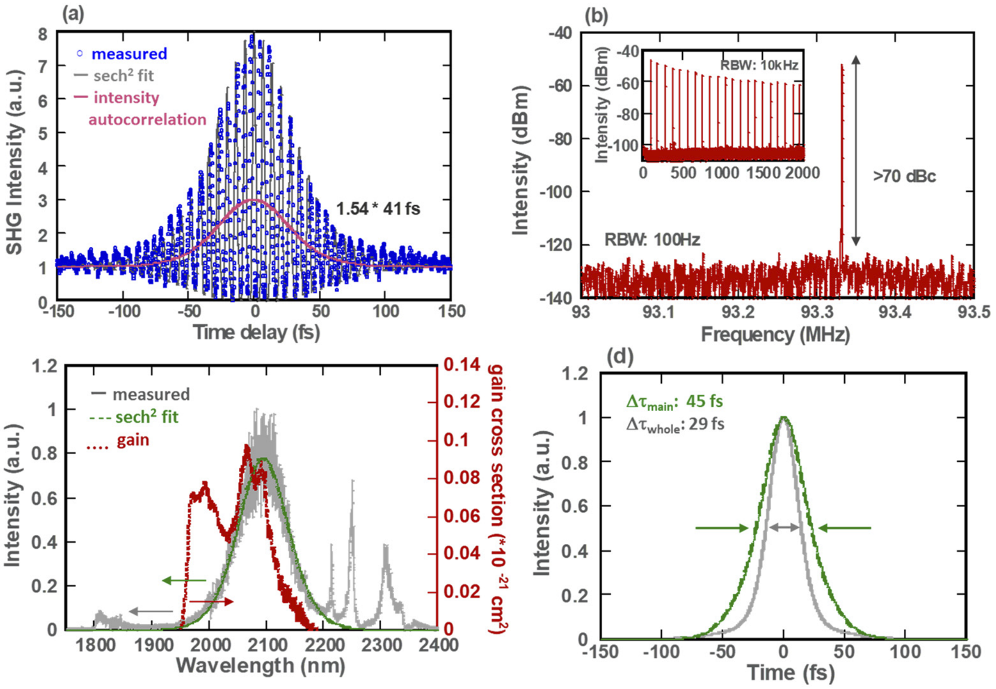

Fig. 6. (a) SHG intensity autocorrelation trace after compression with the 0.5% OC. (b) RF spectrum in 500 kHz and 1 GHz (inset) span range. (c) Optical spectrum of the mode-locked pulses (grey, solid), sech^2 fit (green, dashed) and effective gain cross section (red, dotted). (d) Calculated transform limited pulse with (grey, solid) and without (green, dashed) the 2210-2350 nm spectral component.

gain at these wavelengths. Therefore, a high modulation depth is required in order to sustain mode-locked laser operation without multi-pulsing or the appearance of narrow CW components [15,16]. However, KLM has been shown to be a suitable method to achieve stable mode locking in the anomalous spectral broadening regime, thus overcoming the additional nonlinear losses [14]. In addition, it is worth noting that although the spectra of our lasers are nonlinearly broadened (by Raman processes and/or self-phase modulation), beyond the gain bandwidth, the linearly broadened gain bandwidth of the combined gain media is necessary to sustain mode-locked operation under such large nonlinear spectral broadening effect.

5. Conclusion

In conclusion, we demonstrated a combined gain media KLM laser based on Tm:Lu_2O_3 and Tm:Sc_2O_3. Using a 3% OC, the highest average output power of 316 mW at a pulse duration of 73 fs was achieved. With a 1% OC, pulses as short as 52 fs were obtained. In a high-Q-factor cavity configuration using a 0.5% OC, the shortest pulse duration of 41 fs corresponding to 5.9 optical-cycles was observed assisted by intra-pulse SRS based spectral broadening. To the best of our knowledge, this result represents the shortest pulse duration reported for any Tm -based mode-locked lasers in the 2 µm wavelength range. By combining a Tm:Lu_2O_3 and a Tm:Sc_2O_3 with suitable doping concentrations and pumping conditions, a broadband mode-locked spectrum with a FWHM of more than 100 nm and a wavelength tuning range of nearly 300 nm was obtained. Furthermore, we demonstrated ultrashort pulse generation in the anomalous spectral broadening regime making use of intra-pulse nonlinear processes induced by the use of high-Q factor cavity KLM which enables to exceed the gain bandwidth limitation.

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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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