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High Quality-factor Kerr-lens mode-locked Tm:Sc₂O₃ single crystal laser with anomalous spectral broadening

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We report on a high Quality-Factor Kerr-lens mode-locked Tm:Sc₂O₃ laser operating around 2.1 μm. Using a simple four mirror Z-shaped cavity consisting of an output coupler of 0.5% transmittance, two folding mirrors, and a chirped mirror, pulses as short as 72 fs with an average power of 130 mW were obtained at a center wavelength of 2108 nm. By replacing the 0.5% output coupler with a ~0.3% one, we observed anomalous broadening of the spectrum into the range between ~2250 and 2350 nm where Tm:Sc₂O₃ shows no gain.

Ultra-short pulse 2 μm lasers are attracting great interest for various applications, including wavelength conversion. This does not only include conversion to longer wavelengths from the mid-infrared to the terahertz region by supercontinuum generation¹⁾ or optical-parametric processes²⁻⁴⁾ in non-oxide nonlinear materials. It also covers the shorter wavelength range, e.g. up to the soft X-ray region by high-harmonic generation⁵⁾. In the last decade, there were many efforts to shorten the available pulse duration directly from Tm^{3+} -doped solid state bulk oscillators^{6,7)} and in the last few years, sub-100-fs operation was successfully achieved by the combinations of new gain media and saturable absorbers. As an example, 76 fs pulses were obtained from a single-walled carbon nanotube saturable absorber (SWCNSA) mode-locked $\text{Tm}:\text{MgWO}_4$ laser⁸⁾. Employing $\text{Tm}:\text{Ho}$ co-doped disordered materials, pulses as short as 52 fs were obtained from a GaSb-based semiconductor saturable absorber mirror (SESAM) mode-locked $\text{Tm},\text{Ho}:\text{CaYAlO}_4$ ⁹⁾. Among the Tm^{3+} doped laser materials, $\text{Tm}:\text{RE}_2\text{O}_3$ ($\text{RE}=\text{Sc}, \text{Lu}$ or Y) materials are promising gain media for high power short pulse 2 μm lasers due to their superior spectroscopic, thermo-optical and thermo-mechanical properties¹⁰⁾. Their strong crystal field results in fluorescence bands located at wavelengths beyond 1950 nm, enabling laser operation in the wavelength range between 1.95 μm and 2.2 μm . This operation wavelength range suffers less atmospheric water vapor absorption and ground state absorption, which allows for a broad effective gain bandwidth in mode-locked operation. The high thermal-conductivity of these materials can mitigate undesirable thermal influence during high power laser operation. The available pulse duration from pure $\text{Tm}:\text{RE}_2\text{O}_3$, however, was limited so far. Using an InGaAsSb quantum-well SESAM mode-locked $\text{Tm}:\text{Sc}_2\text{O}_3$ single crystal laser, the pulse duration was limited to 218 fs¹¹⁾, whereas 175 fs pulses were reported from an SWCNSA mode-locked $\text{Tm}:\text{Lu}_2\text{O}_3$ single crystal laser¹²⁾. Very recently, much shorter pulses were generated with mixed ceramics. Pulses as short as 63 fs were demonstrated using an InGaAsSb quantum-well SESAM mode-locked $\text{Tm}:(\text{LuSc})_2\text{O}_3$ mixed ceramic laser¹³⁾ and even shorter pulses of 57 fs durations were demonstrated using a SWCNSA mode-locked $\text{Tm}:(\text{LuY})_2\text{O}_3$ mixed ceramic laser¹⁴⁾. Due to their disordered structure, mixed $\text{Tm}:\text{RE}_2\text{O}_3$ materials exhibit broader gain bandwidths at the cost of a reduced thermal conductivity and often higher scattering losses.

With the aim of the generation of ultrashort pulses at a high average output power, we previously developed a Kerr-lens mode-locked (KLM) $\text{Tm}:\text{Sc}_2\text{O}_3$ laser and obtained pulses as short as 166 fs with an average output power of 0.44 W¹⁵⁾. The main advantages of KLM over other mode locking techniques is, that the Kerr lens acts as a fast saturable absorber enabling a large modulation depth and a broad spectral bandwidth at the same time without

the penalty of large nonsaturable losses. This advantage is crucial to obtain the shortest pulse durations¹⁶⁾. In addition, KLM can be very useful to increase the Q-factor of mode-locked laser cavities. In 2019, Kimura *et al.* reported 22 fs pulse duration from a high Q-factor KLM Yb:CALGO laser with anomalous spectral broadening¹⁷⁾. The spectral broadening overcome the gain bandwidth limitation of Yb lasers. In ¹⁹⁾, This broadening was attributed to Raman-assisted spectral broadening that requires a KLM cavity with a high Q-factor.

Here, aiming to further decrease the pulse durations of 2 μm Tm solid state lasers, we developed a simple 4 mirror high Q-factor Kerr-lens mode-locked Tm:Sc₂O₃ single crystal laser. The cavity consisted of an output coupler, two folding mirrors, and a chirped mirror. With an output coupler of 0.5% transmittance, pulses as short as 72 fs with an average power of 130 mW were obtained at a center wavelength of 2108 nm. By further increasing the cavity Q-factor with an output coupler of less than 0.3% transmittance, we observed anomalous spectral broadening into the range from ~2250 to 2350 nm, where Tm:Sc₂O₃ shows no gain. The phenomenon was similar to the result reported by Kimura *et al.*¹⁷⁾ and should allow further pulse shortening of KLM ultrafast Tm solid state oscillators.

The experimental configuration is shown in Fig. 1. The cavity consisted of an output coupler (OC), two folding mirrors (M1 and M2), and a chirped mirror (CM). The gain medium was a Tm³⁺:Sc₂O₃ single crystal with a doping concentration of 1%, a thickness of 3.7 mm and a group delay dispersion (GDD) of -250 fs² per pass. It was mounted in a water-cooled copper heatsink and placed between two folding mirrors at Brewster's angle. We used in-band pumping at 1611 nm (³H₄ \rightarrow ³F₄) with a home-built Er:Yb fiber MOPA pump source¹⁸⁾. The folding mirror's reflectivity in the wavelength range from 1850 to 2200 nm was >99.8%, and the radius of curvature was 100 mm. The pump light was imaged into the gain medium through one of the folding mirrors, which provided a transmittance of 90% for the pump wavelength. The estimated diameters of the pump laser mode and the cavity fundamental mode at the position of the gain medium were $47 \times 47 \mu\text{m}^2$ and $55 \times 57 \mu\text{m}^2$ (sagittal \times tangential), respectively. The chirped mirror (GDD was -1000 fs² from 2000 to 2200 nm, reflectivity was R>99.9% from 1900 to 2200 nm, UltraFast Innovations LLC) was placed at the end of cavity. We used various OCs with different transmittance, 1% (2000-2300 nm), 0.5% (1900-2100 nm) and ~0.3% (1900-2250 nm, GDD~-200 fs², 2000-2250 nm). A 3 mm thick silicon plate was employed for external pulse compression.

KLM was obtained with all three different OCs. With the 1% OC, the highest average power of 222 mW with a pulse duration of 80 fs was obtained [Fig. 2(a)] The center wavelength and spectral bandwidth were 2141 nm and 65 nm, respectively [Fig. 2(b)] The

repetition rate was 93.8 MHz which is in good agreement with the cavity length [Fig. 2(c)] The maximum pulse energy and peak power were 2.37 nJ and 29.6 kW, respectively. The average output power as a function of the pump power is shown in Fig. 2(d). The KLM was initiated at a pump power of 580 mW by moving the M2. At the onset of KLM the average power doubled, which indicates a large modulation depth. We did not use any hard aperture inside the cavity so the KLM was achieved only by the soft aperture effect.

With the 0.5% OC, the shortest pulse duration of 72 fs with an average output power of 130 mW was obtained [Fig. 3(a)]. The center wavelength and the spectral bandwidth were 2108 nm and 67 nm [Fig. 3(b)] respectively. Due to the narrower bandwidth of the 0.5% OC covering only the range from 1900 to 2100 nm, the center wavelength shifted 33 nm into the shorter wavelength range compared to the 1% OC. The narrow spectral peaks around 2250 nm in Fig. 3(b) are Kelly sidebands. The time-bandwidth product was 0.325, indicating sech^2 pulse shape. Pulse energy and peak power were 1.38 nJ and 19.2 kW, respectively.

Using the 0.3% OC, the broadest spectral bandwidth of 75 nm was obtained at a soliton peak wavelength of 2164 nm (Fig. 4 red dashed curve) with a maximum average power of 50 mW. In addition, we found anomalous spectral broadening into the range between ~2250 and 2350 nm. This region is outside of the gain band of the $^3F_4 \rightarrow ^3H_6$ transition of Tm:Sc₂O₃. The $^3H_4 \rightarrow ^3H_5$ transition could have emission in this range, when pumping with a wavelength of ~800 nm¹⁹⁻²¹⁾ or an upconversion pumping process²²⁾ are applied. However, we are using in-band pumping and the measured fluorescence spectrum under pumping at 1611 nm indicates no fluorescence at wavelengths above 2250 nm (Fig.4 green solid curve). The threshold pump power for this spectral broadening was as low as 500 mW. When the pump power was increased, only the spectral components around 2250-2350 nm grew and the peak intensity around 2164 nm was clipped (Fig. 4). The phenomenon and the spectral shape were similar to the result of the 22 fs Yb:CALGO laser reported by Kimura *et al.*¹⁷⁾. Therefore we attribute the broadening observed here also to Raman assisted spectral broadening. The calculated Raman gain spectrum of our Tm:Sc₂O₃ mode-locked laser is also shown in Fig. 4(black dashed curve). The spectral component around 2250-2350 nm is located inside the Raman gain band but not at the Raman gain maximum position. The mismatch can be explained by the reflectivity and GDD of the cavity. The designed reflectivity and GDD of the chirped mirrors used in our experiment are shown in Fig. 5(a). The reflectivity suddenly decreases at wavelengths longer than 2300 nm and the GDD shows strong high order dispersion above 2250 nm that must be also the origin of the dense Kelly sidebands around 2250 nm. The Raman gain decreases when the group velocity (GV) of the

pump pulse and the Raman pulse do not match, because in that case they do not overlap in the gain crystal during the cavity round trip. So the position of the anomalous spectral broadening component was governed by a balance of GV and reflectivity. It's worth noting that the main sech^2 shape spectral component of our KLM laser was much broader than the fluorescence bandwidth (Fig. 4) and this is the shortest pulse operation mod mode-locked non-disordered Tm solid state laser to our knowledge. Furthermore, assuming the whole spectrum including the Raman gain band at 2250-2350 nm, the corresponding transform limited pulse duration would be ~ 38 fs [Fig. 5(b)] and thus nearly a factor of two shorter than without the 2250-2350 nm spectral component (~ 65 fs).

We also tried to measure the pulse duration by intensity autocorrelation. However, the measured autocorrelation trace with the spectral broadening effect was not stable. We explain the unstable autocorrelation trace as follows: The spectral broadening takes place inside the cavity with the very short crystal length. Under these conditions, the coherence of the whole spectrum can be conserved²³). However, the GDD value of our cavity around 2300 nm was strongly positive and not flat, and therefore the spectral component around 2300 nm was strongly chirped and/or the phase relationship between the main soliton spectral component and the spectral component around 2300 nm was rapidly changing per round trip. Therefore, the Raman band cannot contribute to pulse shortening. This issue can be solved by using a broader bandwidth chirped mirror, which should then result in close to Fourier limited pulse durations utilizing the whole bandwidth of the oscillator. Furthermore, with a proper GDD mirror, the shape of the Raman component should become smoother and the threshold for the spectral broadening should decrease that mitigates the requirement of a high Q-factor cavity and increases an average output power because the effective Raman gain also broadens and increases with a proper GDD mirror. The other requirements to obtain a short pulse duration with this phenomenon would be following: First the cavity should have broad reflectivity bandwidth to fully cover all spectral components. Second the cavity should have a large modulation depth and/or a high Q-factor as the spectral components outside the gain bandwidth of the gain medium does not contribute to stimulated emission and therefore the gain saturation becomes weaker, which can easily lead to multi-pulse operation or the appearance of narrow CW spectral components^{16,24}).

In conclusion, we developed a high Q-factor 4 mirror KLM Tm:Sc₂O₃ laser. With an output coupler of 0.5% transmittance, the shortest pulse duration of 72 fs with an average output power of 130 mW was obtained at a center wavelength of 2108 nm. By further increasing the cavity Q-factor by using the output coupler of less than 0.3% transmittance,

we observed anomalous spectral broadening into the range between 2250 and 2350 nm, where Tm:Sc₂O₃ shows no gain. This phenomenon is attribute to Raman assisted spectral broadening. With proper design of the total cavity dispersion and reflectivity bandwidths, the additional spectral component can contribute to pulse shortening and the spectrum should support sub-40-fs pulse duration.

Acknowledgments

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Figure Captions

Fig. 1. Cavity configuration of 4 mirror Tm:Sc₂O₃ Kerr-lens mode-locked laser

Fig. 2. (a) Autocorrelation trace, (b) spectrum of the mode-locked pulses, (c) RF spectrum, and (d) average power as a function of pump power using a 1% OC.

Fig. 3. (a) Autocorrelation trace, (b) spectrum of the mode-locked pulses using a 0.5% OC.

Fig. 4. Spectra of pulses with a 0.3% OC under the pump power of 0.5 W (blue solid curve) and 1W (red dashed curve) in log scale, and the fluorescence spectrum of Tm³⁺:Sc₂O₃ (green solid curve) and calculated Raman gain (black dashed curve) in linear scale are shown.

Fig. 5. (a) Mode-locked pulse spectrum (black solid curve), reflectivity (red dashed curve) and GDD (blue dotted curve) of the chirped mirrors. (b) Calculated transform limited pulse with (blue dashed curve) and without (red solid curve) the 2250-2350 nm spectral components.

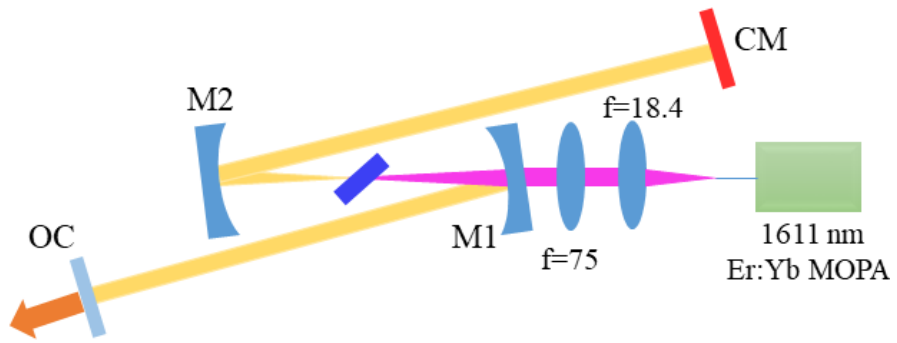


Fig.1.

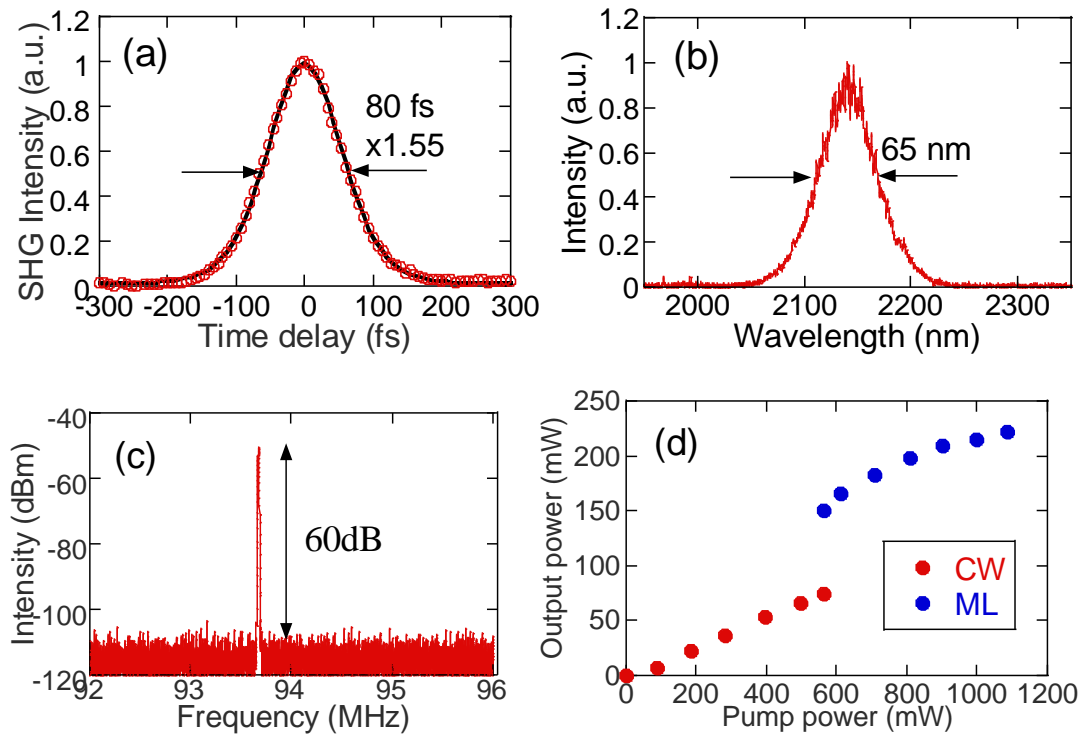


Fig. 2.

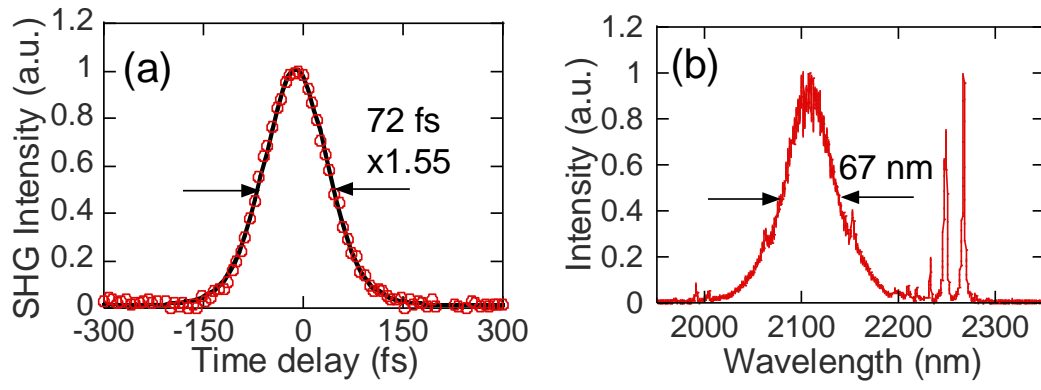


Fig. 3.

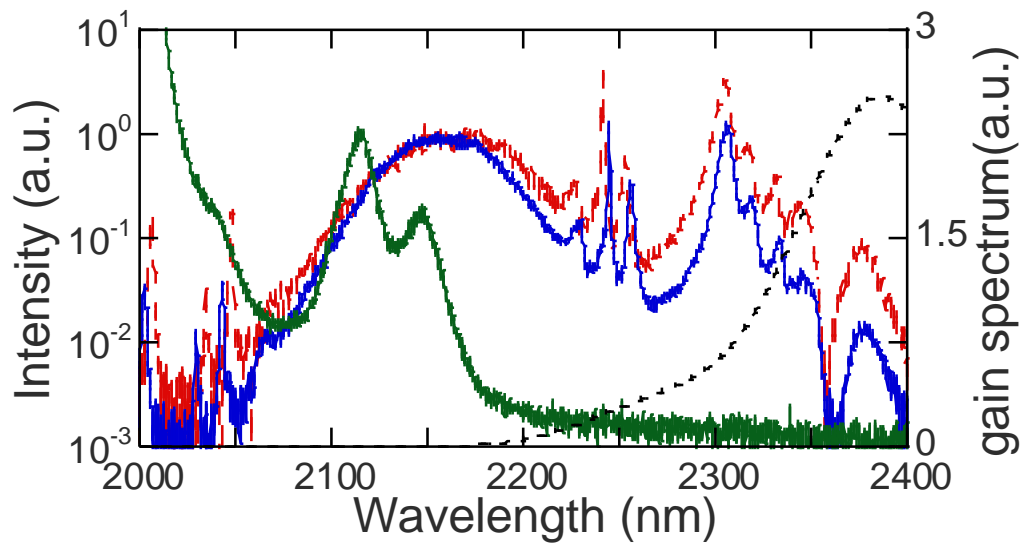


Fig. 4.

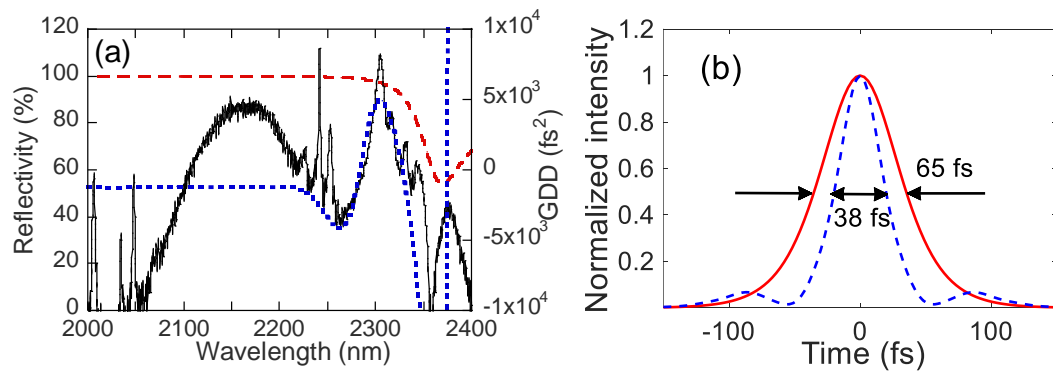


Fig. 5