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In-band pumped Kerr-lens mode-locked Tm,Ho-codoped calcium aluminate laser

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Abstract: We report on a Kerr-lens mode-locked Tm,Ho-codoped calcium aluminate laser with in-band pumping of the Tm ions by a spatially single-mode 1678 nm Raman fiber laser. The structurally disordered $CaGdAlO_4$ host crystal is also codoped also with the passive Lu ion for additional inhomogeneous line broadening. The Tm,Ho,Lu:$CaGdAlO_4$ laser generates soliton pulses as short as 79 fs at a central wavelength of 2073.6 nm via soft-aperture Kerr-lens mode-locking. The corresponding average output power amounts to 91 mW at a pulse repetition rate of ~86 MHz. The average output power can be scaled to 842 mW at the expense of slightly longer pulses of 155 fs at 2045.9 nm, which corresponds to a peak power of ~58 kW. To the best of our knowledge, this represents the first demonstration of an in-band pumped Kerr-lens mode-locked Tm,Ho solid-state laser at ~2 µm.

1. Introduction

Mode-locked (ML) solid-state lasers emitting femtosecond pulses in the 2-µm spectral range at high (~100 MHz) repetition rates present unique opportunities for various applications. Such laser sources are interesting for time-resolved molecular spectroscopy, pump probe studies of semiconductor structures, and characterization of ultrafast detectors. At higher output powers, they can serve as pump sources for synchronously pumped optical parametric oscillators in the mid-infrared (mid-IR) spectral range beyond 5 µm, including frequency comb generation. They can also be used as seed sources for broadband, near-degenerate chirped-pulse optical parametric amplifiers (CPOPAs) pumped near 1 µm, facilitating the generation of high-order harmonics and soft X-rays, or for regenerative laser amplifiers, boosting the single pulse energy at lower

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repetition rates. Finally, femtosecond ML solid-state lasers at 2 µm are feasible for generating mid-IR supercontinuum and terahertz (THz) radiation [1].

Laser emission around 2 µm is commonly achieved using thulium (Tm^{3+}) and holmium (Ho^{3+}) ion based solid-state lasers according to the $^3F_4 \rightarrow {}^3H_6$ (Tm^{3+}) and $^5I_7 \rightarrow {}^5I_8$ (Ho^{3+}) electronic transitions. Lasers employing singly Tm^{3+} (or Ho^{3+}) doped as well as Tm^{3+},Ho^{3+} codoped materials exist. The latter approach is beneficial for several reasons: (i) the Tm^{3+} ion offers more convenient pump wavelengths available from AlGaAs diodes combined with efficient non-radiative energy transfer of the electronic excitation to the Ho^{3+} ion, $^3F_4(Tm^{3+}) \rightarrow {}^5I_7(Ho^{3+})$; (ii) the emission of the Ho^{3+} ion extends beyond 2 µm thus avoiding the structured absorption of water vapors in atmospheric air which may prevent femtosecond pulse generation; (iii) the combined gain bandwidths of both ions may contribute to additional pulse shortening [1]. On the negative side, the substantial heat load caused by energy-transfer upconversion (ETU) from the metastable states of both active ions and energy losses due to the back energy transfer present some challenges for Tm^{3+}, Ho^{3+} codoped materials. It is therefore important to investigate new host materials that combine excellent spectroscopic and thermo-mechanical properties.

The tetragonal (sp. gr. *I*4/*mmm*) calcium rare-earth (*RE*) aluminate Ca*RE*AlO_4 crystals where *RE* denotes Gd or Y, doped with Tm^{3+} and Ho^{3+} ions represent one of the most promising laser materials for generating ultrashort pulses slightly above 2 µm [2]. The Gd- compound is abbreviated in the literature as CALGO and its Y counterpart – as CALYO. The doping of these crystals with laser-active lanthanide (*Ln*) ions is relatively easy due to the presence of the *RE* sites. Both crystals melt congruently which facilitates their growth by the conventional Czochralski (Cz) method [3,4]. The Ca*RE*AlO_4 crystals are optically uniaxial, and their intrinsic birefringence and notable polarization anisotropy of the spectroscopic properties of the dopant ions result in linearly polarized laser emission with weak depolarization losses in high power laser operation (for ***a***-cut crystals). Calcium *RE* aluminates crystallize in a K_2NiF_4-type structure with the Ca^{2+} and RE^{3+} cations statistically distributed over the same Wyckoff site (4*e*, C_{4v}-symmetry) leading to structural disorder [5]. Despite the disordered structure, the Ca*RE*AlO_4 crystals still possess relatively high thermal conductivity with a weak dependence on the active RE^{3+} doping level [6,7].

When co-doped with Tm^{3+} and Ho^{3+} ions, these crystals exhibit inhomogeneous broadening of absorption and emission spectral bands [8]. Due to their broad, flat and smooth gain profiles, they are extremely suitable laser materials for sub-100 fs pulse generation at ~2 µm. Pumped with a continuous-wave (CW) Ti:sapphire laser at 795.3 nm, a Tm,Ho:CALYO laser ML by a SEmiconductor Saturable Absorber Mirror (SESAM) generated 87 fs pulses at 2042.6 nm with an average output power of 27 mW [9]. Shorter pulses of 52 fs at 2015 nm with an average output power of 376 mW were obtained from a SESAM ML Tm,Ho:CALGO laser using the same pump source [8].

The structural disorder characteristic of CALGO and CALYO crystals can be augmented by a compositional disorder, resulting in additional inhomogeneous spectral line broadening. This contributes to smoothening and flattening of the gain profiles of the Tm^{3+} and Ho^{3+} ions. Compositional disorder can be attained e.g. by introducing optically passive Lu^{3+} ions into the CALGO host [10]. Note that the doping level of Lu is at present limited since the stoichiometric compound $CaLuAlO_4$ does not exist. Still the larger ionic radius difference to the Gd host forming ion (compared to Y) means that larger effect can be expected by Lu doping of CALGO compared to CALYO. It was shown that such Tm,Ho,Lu:CALGO crystals feature an additional spectral broadening mechanism due to the compositional disorder leading to red-shifted gain profiles [10]. Using a GaSb-based SESAM as a saturable absorber (SA) and a Ti:sapphire laser tuned to 801.3 nm as a pump source, a Tm,Ho,Lu:CALGO laser delivered soliton-like pulses as short as 46 fs at 2033 nm with an average output power of 121 mW at 78 MHz [11].

In contrast to the traditional pumping of Tm^{3+}-doped laser materials near 800 nm which corresponds to the $^3H_6 \rightarrow {}^3H_4$ transition in absorption, we explore the direct excitation of the Tm^{3+} ion to the $^3F_4$ state ($^3H_6 \rightarrow {}^3F_4$, the so-called Tm in-band pumping). This approach offers a reduction in the heat load by avoiding the non-radiative relaxation path from the $^3H_4$ and $^3H_5$ excited-states of Tm^{3+}. In-band pumping has been employed before for Tm,Ho codoped materials in ~2 µm solid-state lasers in the bulk and waveguide geometries for achieving efficient and power scalable operation in the CW regime [12,13]. More recently also the ML regime was studied using a SESAM [14–16]. In the present work we demonstrate that in-band pumping is feasible also for Kerr-lens mode-locked (KLM) Tm,Ho lasers.

We demonstrate, to the best of our knowledge, the first in-band pumped solid-state KLM laser emitting femtosecond pulses beyond 2 µm, employing a Tm,Ho codoped crystal. The Tm,Ho,Lu:$CaGdAlO_4$ laser generates soliton pulses as short as 79 fs at 2073.6 nm, with an average output power scalable to almost the Watt-level at longer pulse durations of 155 fs centered at 2045.9 nm.

2. Experimental setup

KLM operation was investigated in the X-folded standing-wave cavity depicted in Fig. 1. A 6-mm long rectangular element was cut from a high-quality Tm,Ho,Lu:$CaGdAlO_4$ crystal with measured doping levels of 4.48 at.% Tm^{3+}, 0.54 at.% Ho^{3+}, and 5.51 at.% Lu^{3+} corresponding to a chemical formula of $CaGd_{0.8947}Lu_{0.0551}Tm_{0.0448}Ho_{0.0054}AlO_4$ to enable light propagation along the $\boldsymbol{a}$-axis ($\boldsymbol{a}$-cut). Its 3×3 mm^2 input/output faces were polished to laser-grade quality and anti-reflection coated for both the pump and laser wavelengths. The sample was mounted in a water-cooled copper holder kept at a temperature of 12°C and placed between two dichroic folding mirrors M_1 and M_2 (radius of curvature, RoC = -100 mm).

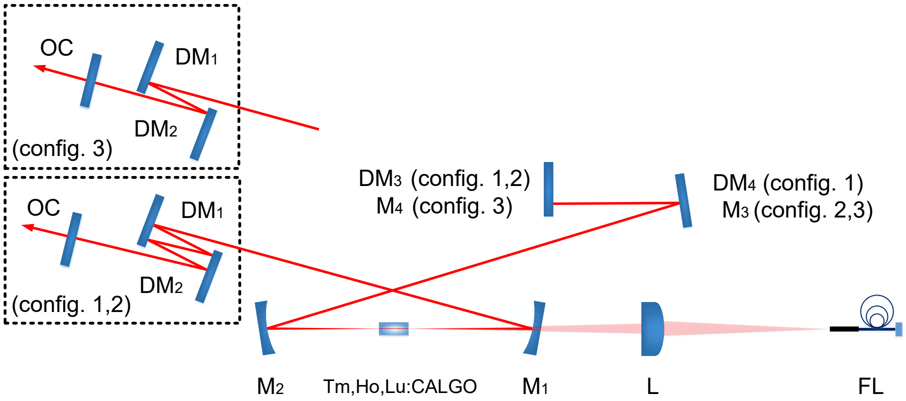

Fig. 1. Schematic of the KLM Tm,Ho,Lu:CALGO laser. FL: Raman-shifted Er-fiber laser; L: spherical focusing lens; M_1 and M_2: concave mirrors; M_3 and M_4: plane mirrors; DM_1 - DM_4: flat dispersive mirrors; OC: plane-wedged output coupler.

We employed a home-made spatially single-mode 6 W Raman-shifted Er-fiber laser emitting unpolarized radiation at 1678 nm [16] as a pump source to address the $^3H_6 \rightarrow {}^3F_4$ Tm^{3+} transition. Its linewidth (full width at the half maximum, FWHM) was 0.8 nm and the maximum incident pump power reached 5.5 W. The pump radiation was focused into the crystal with a spherical focusing lens ($f = 75$ mm), resulting in a beam waist (radius) of 18 µm. Four flat dispersive mirrors (DMs) with negative group delay dispersion (GDD) of -1000 fs^2 per bounce at ~2 µm were used to balance the self-phase modulation (SPM) induced by the Kerr nonlinearity of the laser crystal for soliton-like pulse shaping.

3. Mode-locked laser operation

KLM operation was initially investigated by implementing four DMs providing an overall negative GDD of -11000 fs^2 and an output coupling T_{OC} of 3%, see Fig. 1 (configuration 1). The cavity geometry was adjusted with enlargement of the separation between the two folding mirrors surrounding the laser crystal by a few hundred microns, positioning it close to the edge of the stability region. Stable KLM was then initiated by either knocking on the output coupler (OC) or translating the flat rear mirror (DM$_3$ or M$_3$). Soliton pulses were directly generated from the ML laser, producing an average output power of 842 mW. The absorbed pump power in a single pass was estimated to be 3.72 W. The optical spectrum of the ML laser had a bandwidth (FWHM) of 29.6 nm at a central wavelength of 2045.9 nm, assuming a sech^2-shaped spectral profile, see Fig. 2(a). The autocorrelation trace (AC) based on second harmonic generation (SHG) gave a deconvolved pulse duration of 155 fs (FWHM) assuming a sech^2-shaped temporal intensity profile, as shown in Fig. 2(b). The corresponding time-bandwidth product (TBP) was 0.329, which only slightly exceeded the value for ideal unchirped soliton pulses (0.315). The physical cavity length was ~1.75 m, resulting in a pulse repetition rate of 82.57 MHz. The corresponding peak power amounted to 57.9 kW. The presence of a narrow satellite peak at ~2117 nm is attributed to the limited reflectivity bandwidth of the cavity mirrors and the associated abrupt change in the GDD. This could potentially be improved by using new cavity mirrors with optimized spectral coverage.

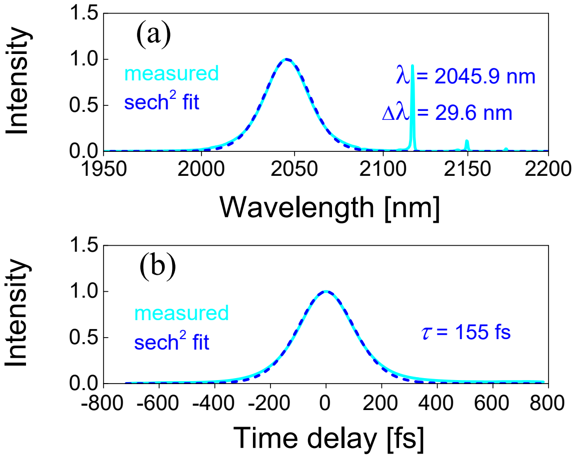

Fig. 2. KLM Tm,Ho,Lu:CALGO laser: (a) optical spectrum and (b) autocorrelation trace for $T_{OC} = 3\%$. *Dashed curves* - sech^2 fits.

By reducing T_{OC} to 1.5% and optimizing the total negative GDD to -9000 fs^2 by a different set of DMs, cf. Fig. 1 (configuration 2), slightly shorter pulses were achieved. Their characteristics are displayed in Fig. 3.

Assuming a sech^2-shaped spectral intensity profile, the emission bandwidth (FWHM) increased to 31.7 nm at a central wavelength of 2040.8 nm, see Fig. 3(a). The recorded SHG-based intensity AC closely matched a sech^2-shaped temporal intensity profile, resulting in an estimated pulse duration of 141 fs (FWHM), see Fig. 3(b). The corresponding TBP was calculated to be 0.322, very close to the Fourier-transform-limit. Under these conditions, the average output power reached 398 mW, for an absorbed pump power of 3.86 W, corresponding to a peak power of 29 kW. A similar satellite peak was observed in this case, too, related again to unmanageable GDD.

The shortest pulse duration was achieved by further reducing the OC transmittance to 0.5% and decreasing the overall negative GDD to -4000 fs^2, see Fig. 1 (configuration 3). The characteristics

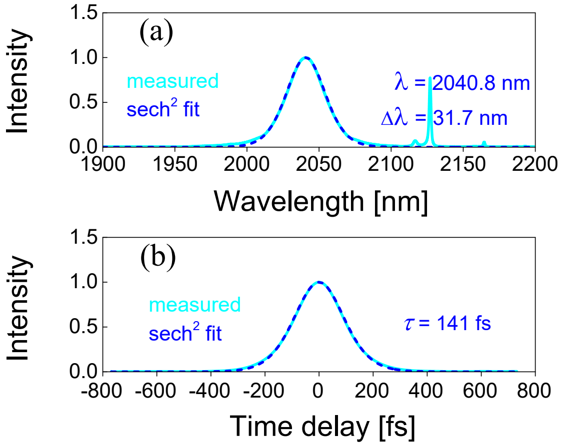

Fig. 3. KLM Tm,Ho:Ca(Gd,Lu)AlO$_4$ laser: (a) optical spectrum and (b) autocorrelation trace for $T_{OC} = 1.5\%$.

of the shortest pulses are shown in Fig. 4. The laser spectrum exhibited a FWHM as large as 58.3 nm, centered at 2073.6 nm, see Fig. 4(a). The pulse duration was assessed from the recorded SHG-based intensity AC trace, which was almost ideally fitted with a sech^2-shaped temporal intensity profile, yielding a deconvolved FWHM of 79 fs. The corresponding TBP was 0.321. Under these conditions, the maximum average output power dropped to 91 mW, for an absorbed pump power of 3.1 W, corresponding to a peak power of 11.8 kW. The narrow satellite peak was more pronounced in this case, and it could not be suppressed by the cavity alignment. The limitation for further pulse shortening in this KLM laser is mainly related to the reflectivity bandwidth of the pump mirror (M$_1$) which exhibits a significant reflectivity reduction well below 2000 nm. We believe that this could be improved in the future with broadband pump mirror as well as optimized GDD management.

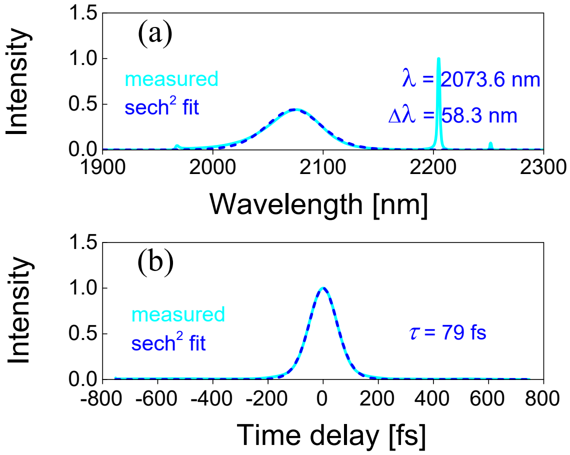

Fig. 4. KLM Tm,Ho:Ca(Gd,Lu)AlO$_4$ laser: (a) optical spectrum and (b) autocorrelation trace for $T_{OC} = 0.5\%$.

The radio frequency (RF) spectra of the shortest pulses were recorded to verify the stability of the ML operation in different frequency span ranges, as shown in Fig. 5. The fundamental beat note located at 85.74 MHz exhibits a high extinction ratio of >72 dBc above carrier, see Fig. 5(a).

The uniform harmonics on a 1-GHz frequency span reveal high stability of the single-pulse CW-ML operation without any Q-switching instabilities, see Fig. 5(b).

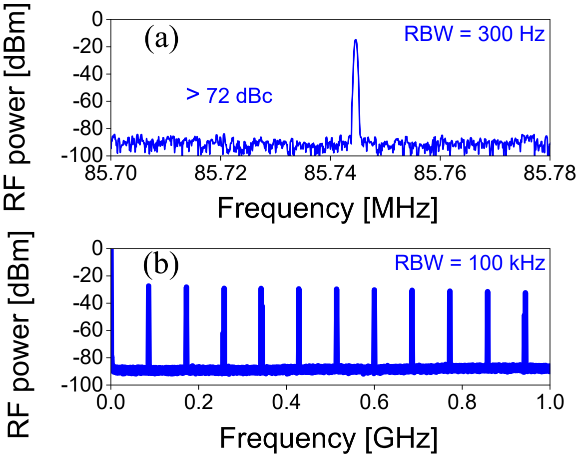

Fig. 5. RF spectra of the KLM Tm,Ho,Lu:CALGO laser: (a) fundamental beat note at 85.74 MHz recorded with a resolution bandwidth (RBW) of 300 Hz, and (b) harmonics on a 1-GHz frequency span recorded with a RBW of 100 kHz.

In Table 1, we summarize the output characteristics of the KLM Tm,Ho,Lu:CALGO laser achieved in the present work. The laser polarization for all the studied OCs was σ and it was naturally selected due to its higher laser gain cross-section. In all aspects the present results were superior to the SESAM ML laser based on the same material and pump source [16].

Table 1. Output Characteristics of the KLM Tm,Ho,Lu:CALGO Laser[a]

T_{OC}, %	P_{out}, mW	P_{abs}, W	$\Delta\tau$, fs	λ_{em}, nm	$\Delta\lambda_{em}$, nm	TPB
3	842	3.72	155	2045.9	29.6	0.329
1.5	398	3.86	141	2040.8	31.7	0.322
0.5	91	3.1	79	2073.6	58.3	0.321

[a] T_{OC} – output coupler transmittance, P_{out} – average output power, P_{abs} – absorbed pump power, $\Delta\tau$ – pulse duration (FWHM), λ_{em} – central emission wavelength, $\Delta\lambda_{em}$ – emission bandwidth (FWHM), TBP – time bandwidth product.

4. Conclusion

To conclude, Tm in-band pumping directly to the $^3F_4$ state is a viable route for attaining power scaling to the Watt-level of Kerr-lens mode-locked Tm,Ho crystalline lasers. It allows to suppress the thermal effects in the gain medium. This pumping scheme can be implemented by using Raman-shifted Er-fiber or short wavelength Tm-fiber lasers. In the present work, a proof-of-principle of this concept of ultrafast laser development is demonstrated with a Tm^{3+} and Ho^{3+} codoped tetragonal calcium rare-earth aluminate crystal $Ca(Gd,Lu)AlO_4$ combining attractive thermo-optical and spectroscopic properties. The latter properties stem from its highly disordered nature: both structural disorder (a random distribution of Ca^{2+} and Ln^{3+} cations over the same Wyckoff site) and compositional one (a partial replacement of Gd^{3+} by Lu^{3+}). Soliton pulses as short as 79 fs were generated at 2073.6 nm with an average output power of 91 mW and a pulse repetition rate at 85.74 MHz. The average output power could be further scaled up to 842 mW at the expense of somewhat longer pulse duration of 155 fs. It shall be emphasized that the crystal ***a***-cut for the present work was selected to utilize the π-polarization for higher absorption of the unpolarized pump beam. Further improvements can be expected employing polarized pump sources. It shall be also outlined that for other crystalline hosts, in-band pumping can have an additional benefit of higher pump efficiency when the absorption of the $^3F_4$ level of Tm is higher compared to the ($^3H_4$) 800 nm range [17].

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Disclosures. The authors declare no conflicts of interest.

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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