$Tm,Ho:KLu(WO_4)_2$ laser mode-locked near 2 µm by single-walled carbon nanotubes

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Abstract: We demonstrate passive mode-locking of a Tm,Ho-codoped crystalline laser operating on the Ho^{3+}-ion transition $^5I_7 \rightarrow {}^5I_8$ near 2 µm using a single-walled carbon nanotube saturable absorber. The $Tm,Ho:KLu(WO_4)_2$ laser emits nearly transform-limited pulses with duration of 2.8 ps at a repetition rate of 91 MHz. The output power amounts to 97 mW.

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

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1. Introduction

In recent years ultrashort pulse crystalline lasers emitting in the 2-μm-spectral range have attracted significant attention because of their potential applications in highly precise surgery [1], free space communication [2], infrared pump-probe spectroscopy and remote sensing [3], material processing of semiconductors and polymers [4], and nonlinear frequency conversion to the mid-infrared range [5,6]. The most popular lasers operating around 2 μm are based on the lanthanide ions Tm^{3+} (Tm) and Ho^{3+} (Ho) [7], which also holds for ultrafast lasers. Compared to the Tm-transition $^3F_4\rightarrow^3H_6$ (slightly below 2 μm), the Ho-ion emission on its $^5I_7\rightarrow^5I_8$ transition is only slightly shifted to longer wavelengths (above 2 μm) but this can have pronounced effect in medical applications due to the variation of water absorption in this range, as well as in nonlinear frequency conversion due to residual absorption in non-oxide nonlinear crystals. Due to the absorption band near 800 nm direct diode-pumping of Tm-lasers is possible because such wavelengths are covered by commercially available GaAlAs laser diodes. Ho-doped lasers are usually excited by energy transfer through co-doping with

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Tm-ions profiting from quantum efficiency up to two via cross relaxation [8] or by direct in-band pumping with Tm fiber or bulk lasers [9].

The first rare-earth mode-locked lasers with sub-ps pulse duration near 2 μm were based on Tm-doped fibers. In 1995, Nelson et al. generated 500-fs pulses [10] using the nonlinear polarization rotation approach. Shortly afterwards, Sharp et al. demonstrated 190-fs pulses using a semiconductor saturable absorber mirror (SESAM) [11]. Mode-locking based on saturable absorption had not been demonstrated with Tm-, Ho-, or Tm,Ho-doped crystalline laser media until 2009. One possible reason could be the water vapor absorption. The broad and smooth gain spectra of most Tm-doped crystalline laser media permit tunable mode-locked operation or generation of sub-ps pulses in the spectral range between 1.8 and 2.1 μm (depending on the host) with a gain maximum typically between 1.90 and 1.95 μm [7]. However, the strong water vapor absorption for wavelengths shorter than 2.0 μm can prevent continuous tunability and broadband mode-locking unless elaborated purging (e.g. nitrogen) or other kinds of cavity evacuation is performed. Hence, two approaches are applied. The first one is shifting the laser emission spectrum of Tm-doped media still displaying a noticeable gain in the long wavelength wing beyond 2 μm at the expense of lower gain and laser efficiency. The second approach overcomes this limitation using Ho-, or Tm,Ho-codoped crystalline laser media with highest stimulated emission cross section above 2 μm.

Starting in 2009, different kinds of saturable absorbers (SA) have been utilized to mode-lock a large number of gain media. Using SESAMs for Tm- [12–15], Tm,Ho- [15–19] and Ho-doped [20] bulk materials ultrashort pulse generation was demonstrated, whereas all carbon-nanotube SA and graphene-SA mode-locking was realized only with Tm-doped hosts [21–25].

The SESAM [26] is a well-established device for ultrashort pulse lasers, providing a robust intracavity mode-locking mechanism. However, SESAMs only offer useful saturable absorption in a narrow spectral range and often require sophisticated manufacturing technology [27]. As an alternative, single-walled carbon nanotubes (SWCNTs) and graphene have been introduced because of their unique physical and optical properties [28,29]. Both are characterized by fast recovery time and can be fabricated by relatively simple processes [30].

In this work we apply a carbon nanostructure-based SA for mode-locking of the 2-μm Ho-ion transition $^5I_7 \rightarrow {}^5I_8$ for the first time to our knowledge. The laser material is co-doped monoclinic Tm,Ho:$KLu(WO_4)_2$ and the SA is based on SWCNTs.

2. Experimental setup

As active medium served a Tm,Ho:$KLu(WO_4)_2$ (Tm,Ho:KLuW) crystal [31,32], which is the first implementation of this material for mode-locked operation. Such monoclinic potassium double tungstates, doped with active rare-earth ions, are established as promising solid-state laser materials. In this class of biaxial crystals, the gain bandwidths are very broad and the absorption and emission cross-sections are very high for selected polarizations. In particular, the KLuW crystal is very suitable as host for Yb, Tm and Ho-ions and provides very high laser efficiencies [31]. The Tm,Ho:KLuW is grown by the Top Seeded Solution Growth technique with 5at.% Tm^{3+} and 0.5at.% Ho^{3+}-concentration in the solution.

The design of the mode-locked laser is based on a 1.6 m long X-type linear cavity (Fig. 1). It consists of four curved mirrors (M1, M2, M3 and M4) and a plane output coupler. The calculated cavity waist radii are 32 μm both at the position of the laser crystal and the SA. The latter is placed at Brewster angle near the second cavity waist formed by mirrors M1 and M2. The 3-mm long Tm,Ho:KLuW crystal is mounted in a copper holder for active water cooling and is oriented for beam propagation along the N_g crystallo-optic axis and for polarization along the N_m axis. Positioned at Brewster angle in the central folding section the crystal is pumped by a Ti:sapphire laser emitting at 802 nm. The pump beam is focused to a radius of ~30 μm. The mirrors M3 and M4 are highly transmitting for the pump radiation and the curved mirror M5 acts as a back-reflector for the unabsorbed pump. The single pass

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absorption of the Tm,Ho:KLuW active element in the lasing state was 87% and estimated to ~95% for double-pass pumping.

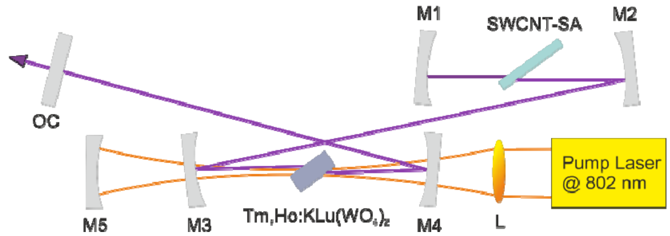

Fig. 1. Layout of the Tm,Ho:KLu(WO$_4$)$_2$ laser. L: focusing lens (f = 75 mm); M1: concave mirror with radius of curvature (ROC) = 50 mm; M2, M3, M4 and M5: concave mirrors (ROC = 100 mm); M1, M2, M3, M4: highly reflective around 2.06 µm and highly transmitting around 800 nm; M5: highly reflective around 800 nm; OC: output coupler.

The SA is based on arc-made single-walled carbon nanotubes, layered on a 1-mm thick quartz substrate by the spin coating technique. More about the fabrication can be found in [30]. The absorption bands are spectrally shifted to longer wavelengths compared to the HiPCO (high-pressure CO conversion decomposition) SWCNT-based SA, so that the E_{11} interband transition can be applied for wavelengths longer than 2 µm [23]. The linear absorption of the SWCNT-SA at 2.06 µm amounts to 1.4% and the measured recovery time for such type of SA delivered a fast response of the order of 1 ps [33]. The expected values for the saturation fluence and the modulation depth are <10 µJ/cm^2 and <1%, respectively [30]. The samples are optimized with respect to low scattering loss by reducing SWCNT knotting and interweaving into one another compared to our previous reports [33].

3. Results

At first the laser cavity is optimized in the continuous-wave (CW) regime without introducing the SWCNT-SA. Using a 1.5% transmission output coupler (T_{OC}) the laser delivers 187 mW of output power for 900 mW of absorbed pump power. Figure 2 shows the measured average output power versus the absorbed pump power of the laser containing the SWCNT-SA and the same output coupler. The laser threshold corresponds to ~150 mW of absorbed pump power and starts in the CW mode. Mode-locked operation disturbed by some Q-switching instabilities is observed in the pump power range between 500 mW and 700 mW detected by a small pedestal of the first beat note of the recorded radio frequency spectrum. The threshold for the self-starting CW mode-locked (ML) regime is 68 mW of output power obtained at an absorbed pump power of 700 mW. The ML threshold intracavity pulse energy of 50 nJ satisfies the condition for suppression of Q-switching instabilities, derived by Hönninger et al. [34], with the emission cross section of Tm,Ho:KLuW of 1.19 × 10^{-20} cm^2 at 2060 nm [32]. The maximum output power in ML operation is 97 mW at an absorbed pump power of 970 mW resulting in a slope efficiency of 12% which is about 10% larger than in the CW regime due to the saturated loss of the SA (Fig. 2).

The pulse duration (FWHM) at maximum output power is 2.8 ps, derived from the measured autocorrelation trace, assuming a sech^2-pulse shape (Fig. 3). The measured optical spectrum has a FWHM of 1.6 nm and the laser wavelength is centered at 2059 nm (inset Fig. 3). The time-bandwidth product amounts to 0.32 corresponding to almost transform-limited pulses. It should be noted that no significant variations of the optical spectrum (FWHM), respectively pulse duration, could be achieved when two intra-cavity CaF_2-prisms are introduced between M4 and the OC in the cavity. Self-starting and stable ML is also achieved using T_{OC} = 0.2%, 0.5%, 3% and 5%. The shortest pulse duration of 2.4 ps is obtained applying T_{OC} = 0.5% with an output power of 43 mW. The maximum output power of 107 mW is achieved for T_{OC} = 3% at the expense of slightly longer pulses of 5.3 ps duration.

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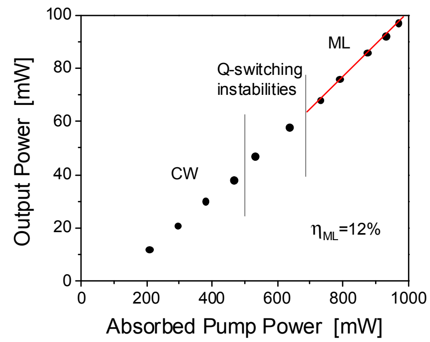

Fig. 2. Input–output characteristics of the Tm,Ho:KLu$(WO_4)_2$ laser mode-locked by a SWCNT-SA (T_{OC} = 1.5%). Red line: slope efficiency (η) in the mode-locked regime (linear fit).

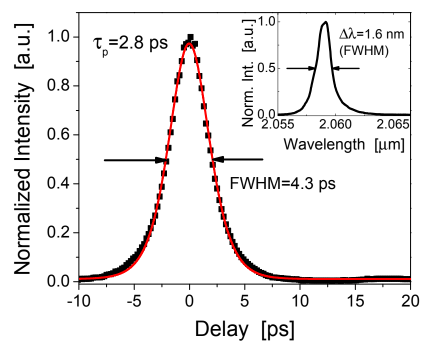

Fig. 3. SWCNT-SA mode-locked Tm,Ho:KLu$(WO_4)_2$ laser: Autocorrelation curve (black dots), fit assuming a sech^2-pulse shape (red line), and corresponding optical spectrum (inset).

Figure 4 shows the measured radio-frequency spectrum of the SWCNT-SA mode-locked Tm,Ho:KLuW laser output using an RF spectrum analyser (Rhode&Schwarz). It contains the fundamental beat note at 90.6 MHz, see Fig. 4(a), recorded with a resolution bandwidth of 1 kHz, and 1 GHz wide-span measurement, see Fig. 4(b). The high extinction ratio of 67 dBc and the absence of spurious modulation indicate very stable ML operation.

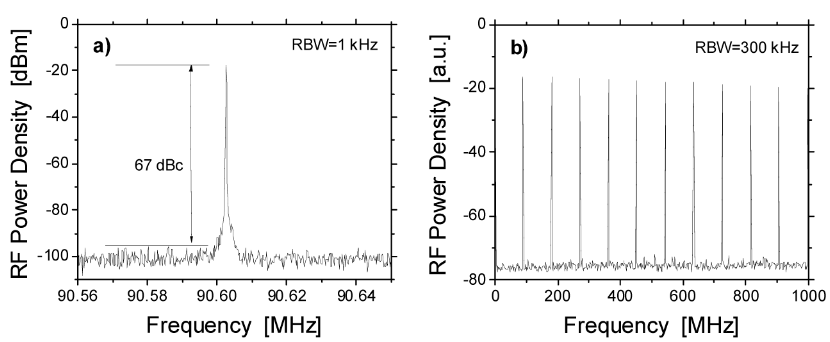

Fig. 4. Radio frequency-spectrum of the SWCNT-SA mode-locked Tm,Ho:KLu$(WO_4)_2$ laser: (a) fundamental beat note, (b) 1 GHz wide-span. (RBW: resolution bandwidth).

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4. Conclusion

In conclusion, a Tm,Ho:KLu$(WO_4)_2$ laser operating at 2.06 μm was mode-locked using a SWCNT-SA. We demonstrate carbon nanostructure-based mode-locking of the 2-μm Ho-transition for the first time. A stable pulse train was generated with average output power of 97 mW and almost Fourier-limited pulse duration of 2.8 ps at a repetition rate of 91 MHz.

A potential reason for not achieving sub-ps pulse durations as demonstrated with Tm:KLuW [23] and Tm:Lu_2O_3 [21] when operating the SWCNT-SA mode-locked laser around 2.05 μm is the ~4 times larger emission cross section of Tm,Ho:KLuW [32]. As a consequence a higher modulation depth of the SWCNT-SA is required to support even shorter pulses. This assumption is supported by applying InGaAsSb SESAM structures, typically characterized by a higher modulation depth, for mode-locking other Tm:Ho-codoped double tungstates. The Tm,Ho:KY$(WO_4)_2$ [16] and Tm,Ho:NaY$(WO_4)_2$ lasers [17] delivered pulses with sub-ps duration and output powers around 150 mW. Also GaSb-based SESAMs have been applied for mode-locking of Tm,Ho:YAG [18] and Tm,Ho:YAP [19] lasers with maximum output power reaching 285 mW, however minimum pulse durations were longer than 20 ps.

Compared to the other mode-locked Tm,Ho-codoped lasers, the presented initial results with SWCNT-SA are quite encouraging. Applying SWCNT-SAs designed for slightly higher modulation depth (e.g. higher concentration of SWCNTs) in a next step one can expect further pulse shortening in the sub-ps range with Tm,Ho:KLuW.

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