Passive mode-locking of a Tm-doped bulk laser near 2 μm using a carbon nanotube saturable absorber

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Abstract: Stable and self-starting mode-locking of a $Tm:KLu(WO_4)_2$ crystal laser is demonstrated using a transmission-type single-walled carbon nanotube (SWCNT) based saturable absorber (SA). These experiments in the 2 μm regime utilize the E_{11} transition of the SWCNTs for nonlinear saturable absorption. The recovery time of the SWCNT-SA is measured by pump-probe measurements as ~1.2 ps. The mode-locked laser delivers ~10-ps pulses near 1.95 μm with a maximum output power of up to 240 mW at 126 MHz repetition rate.

OCIS codes: (140.4050) Mode-locked lasers; (140.5680) Rare earth and transition metal solid-state lasers; (160.4236) Nanomaterials; (160.4330) Nonlinear optical materials

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1. Introduction

Solid-state lasers operating in the 2 μm eye-safe spectral range are interesting for applications in spectroscopy, remote sensing, photo-medicine, optical communications, and metrology [1,2]. Additionally, mode-locked lasers near 2 μm appear particularly useful for time-resolved molecular spectroscopy, as pump sources for synchronously-pumped OPOs operating in the mid-IR above 5 μm, as seeders of degenerate chirped-pulse optical parametric amplifiers pumped near 1 μm for the generation of high-harmonics and soft X-rays, and for IR supercontinuum or THz generation. Both, Tm^{3+} and Ho^{3+}-doped materials operate as three-level systems in this wavelength range. Yet, the advantage of the Tm-laser is the possibility of direct diode pumping with AlGaAs diodes whereas Ho^{3+} normally requires co-doping, e.g. with Tm^{3+}. Moreover, the Ho^{3+} gain profile exhibits narrow spectral features. Recently, numerous Tm^{3+}-doped materials showed high efficiency and output power in the continuous-wave and Q-switched laser operation [3,4]. Despite almost two decades of efforts, both, Tm and Ho bulk lasers have been mode-locked only by active amplitude modulation. Acousto-optic modulation produced 45 ps long pulses at 2.02 μm in Cr:Tm:YAG [5] and 35-ps pulses at 2.01 μm with Tm:YAG [6]. Nonlinear polarization rotation [7,8], semiconductor saturable absorber mirrors (SESAMs) [9], and carbon nanototube saturable absorbers [10,11] have been successfully used to mode-lock Tm-fiber lasers, but, to the best of our knowledge, Tm bulk lasers with their higher sensitivity to cavity losses have never been passively mode-locked so far. Output powers of mode-locked Tm-fiber lasers appear intrinsically limited, with reported values in the mW range or below. Only very recently 178 mW were achieved in a double clad fiber [8]. Here we demonstrate stable and self-starting mode-locking of a bulk $Tm:KLu(WO_4)_2$ (Tm:KLuW) laser using a single-walled carbon nanotube (SWCNT) based saturable absorber (SA).

SWCNT-SAs exhibit unique properties, in particular intrinsically fast saturable absorption [12], which makes them an interesting alternative to SESAMs. While SESAMs have to be fabricated using sophisticated epitaxial technology [13], SWCNT-SAs can be manufactured with relatively simple technology and at low cost. The first bulk laser that was passively mode-locked by a SWCNT-SA operated near 1.5 μm and was based on Er/Yb:glass [14]. Recently, we demonstrated passive mode-locking of a Cr:forsterite laser at 1.25 μm [15] and also of Yb-doped double tungstate lasers such as Yb:KLuW in the 1 μm range [16]. Since the absorption band of the SWCNT is controllable by varying the tube diameter and its

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chirality, SWCNT-SAs are readily applicable within a broad spectral range, extending roughly from 1 µm to 2 µm in the near-infrared. This strongly contrasts SESAMs in this range, which require transition through several material systems and which have not reached the maturity level of GaAs-based technology applied near 1 µm, yet.

2. Characteristics of SWCNT-SA

The SWCNT-SAs used in the present work are of the same type as those used for bulk solid-state laser mode-locking in the 1 µm spectral range [16]. However, while the absorption band around 1 µm corresponds to the E_{22} transition of SWCNTs, the E_{11} transition can be utilized for mode-locking of Tm- or Ho-doped laser materials at 2 µm.

SWCNTs synthesized by the arc-discharge method (Iljin Nanotech) were used as the starting material. Vacuum-dried SWCNTs with a concentration of ~0.1 mg/ml were dispersed in dichlorobenzene (DCB) via ultrasonication. This SWCNT dispersion and a polymethyl methacrylate (PMMA) solution were subsequently mixed at the volume ratio of 1:1. After stirring overnight, the SWCNT-SA film was deposited on a quartz substrate using spin coating. The coated film thickness was measured to be approximately 300 nm by an Alpha-step surface profiler. The detailed manufacturing process is described elsewhere [17]. Figure 1 clearly shows the two absorption bands (first interband transition E_{11} with peak absorption at about 1.8 µm and second transition E_{22} at 1 µm) in the transmission spectrum of the SWCNT-SA used in the experiments.

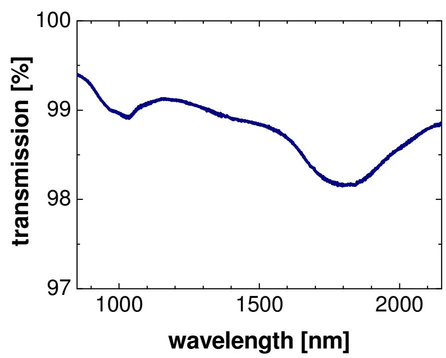

Fig. 1. Transmission spectrum of the transmission-type single-walled carbon nanotube saturable absorber (SWCNT-SA). Measurement has been corrected for Fresnel losses.

In order to understand the dynamic transmission, the resonant response of the saturable absorption was characterized by pump-probe measurements. For this purpose, sub-200 fs pulses from a tunable optical parametric oscillator (OPAL, Spectra Physics) operating at 1.92 µm were focused on the sample, which yields pump pulse fluences of about 10 µJ/cm^2. The recovery characteristics of the transmitting SWCNT-SA were determined in a non-collinear cross-polarized pump-probe measurement setup.

The measured dynamic transmission response of the saturable absorber at 1.92 µm and a fit to the data are shown in Fig. 2. The analysis reveals that the response is well described by a biexponential decay with one nearly instantaneous component and a second slower component with a decay time of about 1 ps. This is at least comparable to the fastest response reported for GaAs based SESAMs. Our measurement indicates a relative weight of the quasi-instantaneous response of about 25%. The measured recovery times of the transition are nearly identical to such previously reported for the E_{22} transition [17].

We have not been able to characterize saturation fluence and modulation depth of the SWCNT-SAs near 2 µm, yet. Nevertheless, we expect these values also to be very similar to the parameters of the E_{22} transition of the same SWCNTs [17]. Recently, we have confirmed

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this assumption by using the same E_{11} transition of HiPCO (high-pressure CO conversion) SWCNTs for bulk laser mode-locking near 1.3 μm [15], i.e., saturation fluence: < 10 μJ/cm^2 and modulation depth: < 0.5%.

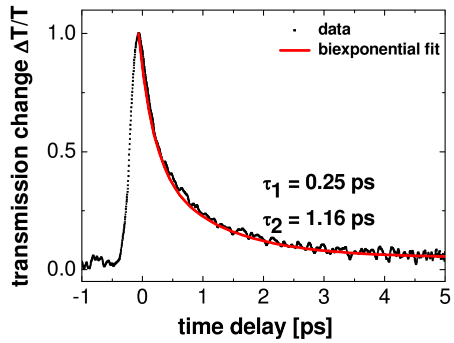

Fig. 2. Nonlinear response of the transmission-type SWCNT-SA at 1.92 μm. The solid curve shows a biexponential fit to the data.

3. Mode-locking of Tm:KLuW laser

In the mode-locking experiments we pumped the Tm:KLuW laser with a continuous-wave (cw) Ti:sapphire laser tuned to 802 nm. The laser set-up was a modified version of the one described in [18], with an additional folding mirror of -10 cm radius of curvature (RC) and a RC = -5 cm end mirror to produce a second cavity waist for the SWCNT-SA. The measured beam waist was about 30 μm and the transmission-type SWCNT-SA was inserted at Brewster angle close to this waist. The output coupler terminating the other cavity arm had a transmission of 1.5%. The anisotropic host material, KLuW, was selected not only because it previously showed highly efficient laser operation but also because in comparison to the other Tm hosts it exhibits the highest interaction cross-sections (for polarization parallel to the $\boldsymbol{N_m}$-axis) while the fluorescence lifetime is relatively short. Such properties are considered beneficial for stabilizing the passive mode-locking process against Q-switching. The KLuW sample was 3 at. % Tm-doped (concentration $n = 2.4\times10^{20}$ cm^{-3} measured in the crystal). It was 2.92 mm thick with an aperture of 3×3 mm^2. The active element was cut for propagation along the $\boldsymbol{N_g}$ principal optical axis and used polarization parallel to the $\boldsymbol{N_m}$ optical axis. It was uncoated and positioned at Brewster angle in the vicinity of the pump focus, in the second folding section of the cavity. Its Cu holder was supplied with water cooling.

Exploiting the versatility of transmittive saturable absorbers stable Q-switching and stable mode-locking without any Q-switching were achieved using the same SWCNT-SA. We could switch between the different modes of operation by slightly realigning the cavity, changing the effective beam waist on the transmittive SWCNT-SA. Using the above discussed saturation fluence and modulation depth of the SWCNT-SA, the measured waist size of 30 μm on the absorber and the saturation fluence of Tm:KLuW at 1950 nm [18], we estimate stable mode-locked operation [19] for output powers $P > 10$ mW. Given that this is close to the threshold for mode-locking, the laser is practically unconditionally stable against Q-switched mode-locking, which is explained by the small saturation fluence of the SWCNT-SA as well as its small modulation depth. However, increasing the spot size on the absorber to > 90 μm, Q-switched mode-locking is expected for the entire range of output powers obtainable with our pump source.

First we studied Q-switched operation. In this regime, the laser operated at ~33 kHz repetition rate, and an average output power of 170 mW was obtained. As shown in Fig. 3, the period of the recorded pulse train was substantially shorter than the 1.34 ms fluorescence lifetime of Tm:KLuW.

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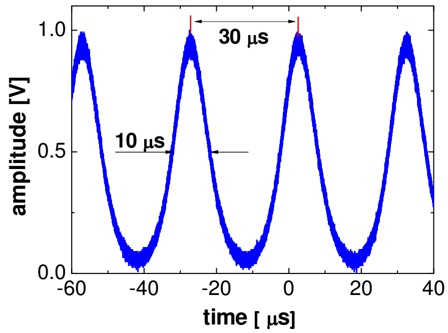

Fig. 3. SWCNT-SA Q-switched Tm:KLuW laser: steady-state operation.

Only cavity alignment and, in particular, optimization of the position of the SWCNT-SA in the direction of the beam propagation close to the waist allowed to achieve stable mode-locking rather than Q-switching. Once this alignment was found, mode-locking was self-starting and stable over hours. When reducing the pump power almost down to the laser threshold, mode-locking could be sustained with only slight cavity realignment. Neither degradation nor damage of the SWCNT-SA was observed after several weeks of operation.

The duration of the mode-locked pulses was measured with an intensity autocorrelation in a 6 mm-thick type-I β-barium borate (β-BBO) crystal. Figure 4(a) shows the autocorrelation trace recorded at an output power of > 200 mW. Assuming a $sech^2$-shaped pulse, its duration is 9.7 ps (FWHM). We recorded the laser spectrum with a spectrometer at a spectral resolution of 0.2 nm. The spectrum was centered near 1944 nm (Fig. 4(b)) and exhibited a spectral bandwidth (FWHM) of 0.45 nm. This corresponds to a time-bandwidth product of 0.347, which is close to the Fourier limit for a $sech^2$ pulse.

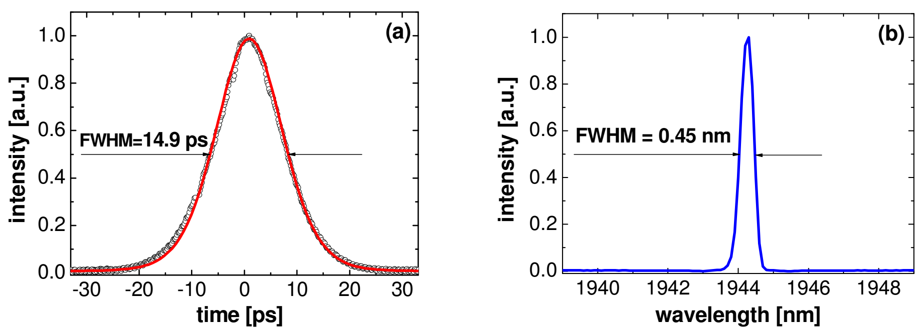

Fig. 4. SWCNT-SA mode-locked Tm:KLuW laser: (a) autocorrelation trace and (b) spectrum.

The radio-frequency (RF) spectrum of the SWCNT-SA mode-locked Tm:KLuW laser is shown in Fig. 5. Measured at a resolution bandwidth of 1 kHz and a 400 kHz span, the fundamental beat note at 126.03 MHz [Fig. 5(a)] displays an extinction ratio of > 60 dB above carrier. As further evidence for stable cw single-pulse operation without Q-switching, Fig. 5(b) depicts a wide-span RF measurement.

Figure 6 shows the measured average output power versus the incident pump power in the mode-locked regime. The maximum output of 240 mW was obtained for a pump level of 1.86 W at 802 nm pump wavelength. Pump absorption in the crystal amounts to about 85%. Higher average power was readily generated with increasing pump power, however, the laser became unstable, and the emerging multiple pulsing could not be suppressed easily.

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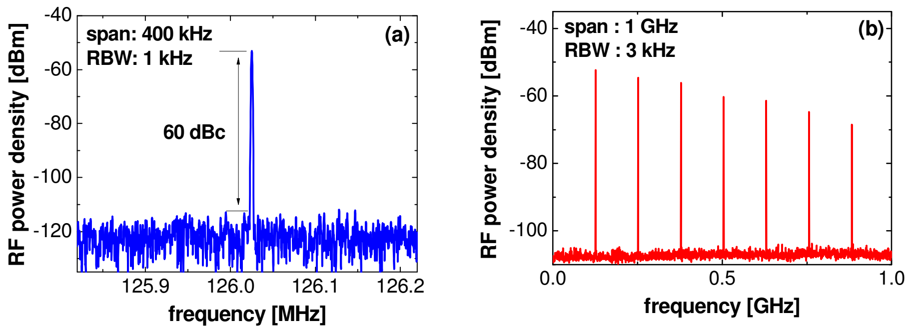

Fig. 5. RF spectrum of the mode-locked Tm:KLuW laser without indications of Q-switching: (a) First beat note at 126.03 MHz. (b) Wide-span RF spectrum.

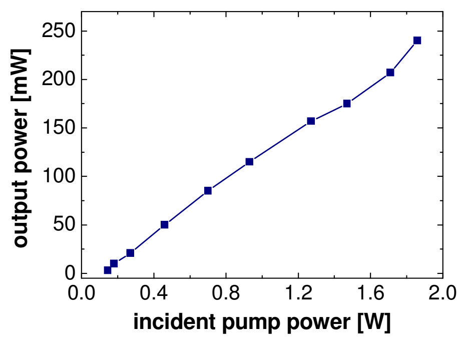

Fig. 6. Mode-locked Tm:KLuW laser: average output power versus incident pump power, measured with 1.5% output coupler transmission.

4. Conclusions

For the first time to our knowledge, passively mode-locked operation of a rare-earth doped bulk laser near 2 µm was demonstrated. A transmission-type SWCNT-SA, utilizing the E_{11} electronic transition of SWCNTs for saturable absorption, served as the passive mode-locking device for a three-level Tm-laser based on the KLuW host. Bandwidth-limited pulses with duration of ~10 ps at 1.94 µm were generated in a self-starting and stable single-pulse regime. Average powers up to 240 mW at a repetition rate of 126 MHz were obtained. It is interesting to note that one and the same SWCNT-SA enabled pure Q-switching at ~33 kHz as well as clean mode-locking. Previous cw laser experiments on Tm:KLuW indicated a wide tuning range sufficient to host pulses of sub-50 fs duration [18]. Consequently, further experiments will be directed towards exploitation of this bandwidth potential for femtosecond pulses. The combination of Tm:KLuW and SWCNT-SAs therefore appears promising for passively mode-locked all-solid-state femtosecond oscillators in the emerging wavelength range around 2 µm.

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