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67-fs pulse generation from a mode-locked Tm,Ho:CLNGG laser at 2083 nm

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Abstract: We report on a mode-locked Tm,Ho:CLNGG laser emitting in the 2 µm spectral range using single-walled carbon nanotubes (SWCNTs) as a saturable absorber (SA). Pulses with duration of 98 fs are generated at 99.28 MHz repetition rate with an average output power of 123 mW, yielding a pulse energy of 1.24 nJ. Using a 0.5% output coupling, pulses as short as 67 fs, i.e., 10 optical cycles, are produced after extracavity compression with a 3-mm-thick ZnS plate.

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1. Introduction

Mode-locked lasers operating in the femtosecond regime are attractive for applications in time-resolved optical spectroscopy, high-field physics, and light-matter interaction. With a central wavelength in the 2 µm spectral range they can be used as synchronous pump sources for generation of widely tunable mid-IR pulses at high repetition rates from optical parametric oscillators [1] or as seed sources for broad-band near-degenerate chirped pulse optical parametric amplification [2]. Mode-locked bulk solid state lasers offering much lower excess noise compared to femtosecond fiber oscillators are more suitable for further power scaling with carrier-envelope phase (CEP) stabilization [3]. Tm^{3+} and/or Ho^{3+} doped materials are employed in such solid-state lasers [4–9]. In comparison, Ho-lasers with slightly longer emission wavelength (to avoid the structured water vapor absorption) and larger emission cross-section [10] are preferable for the above mentioned applications. To date, by exploiting inhomogeneous spectral broadening of the emission of co-doped disordered host materials, 191-fs pulses at 2060 nm have been generated with a semiconductor saturable absorber mirror (SESAM) mode-locked tetragonal Tm,Ho:$NaY(WO_4)_2$ laser [5]. Most recently, we demonstrated the first sub-100-fs (87-fs) pulse generation with a SESAM mode-locked tetragonal Tm,Ho:$CaYAlO_4$ (CALYO) laser [7].

$Ca_3Nb_{2-x}Ga_{3+x}\square_yO_{12}$ (shortly CNGG, the □ symbol represents the cationic vacancies), exhibits a disordered structure with cubic symmetry (optically isotropic) and is another prominent example for inhomogeneous spectral line broadening of the absorption and

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emission of the active trivalent dopant ions [11,12]. By further doping with univalent alkali cations such as Li^+, i.e., CLNGG, the unwanted cationic vacancies serving for charge compensation can be eliminated, thereby improving its optical quality and laser performance. To date, 900-fs pulses at 1061 nm have been generated from a SESAM mode-locked Nd:CLNGG laser [13], and 55-fs pulses at 1051.5 nm have been reported using a Yb:CLNGG crystal [14]. In the 2 µm spectral range, Tm:CLNGG has been studied before [15,16] and the shortest pulse duration reported amounted to 78 fs at 2017 nm [6]. However, laser operation of singly Ho- or Tm,Ho co-doped CLNGG has never been reported. On the other hand, mode-locking of Tm,Ho:CNGG has been recently investigated achieving sub-80-fs pulses after external compression [8]. Based on the above considerations, mode-locking of disordered Tm,Ho co-doped CLNGG laser is expected to be one of the promising routes towards few-optical-cycle pulse generation in the 2 µm spectral range.

In this letter, we investigated the continuous-wave (CW), wavelength tuning and passive mode-locking of the Tm,Ho:CLNGG laser using a single-walled carbon nanotube (SWCNT) saturable absorber (SA), generating pulse durations equivalent to 10 optical cycles at 2083 nm.

2. Experimental details and discussion

A scheme of the tunable and passively mode-locked Tm,Ho:CLNGG laser is shown in Fig. 1. A wavelength tunable Ti:Sapphire laser with near-diffraction-limited beam quality was used as the pump source. The pump wavelength was selected at 796 nm for matching optimal crystal absorption. A maximum pump power of 3.3 W was applied. An anti-reflection (AR)-coated lens (f = 70 mm) was used to focus the pump beam onto the crystal with a beam radius of 30 µm (normal incidence). The dimension of the Tm (2.34 at.%), Ho (0.54 at.%) co-doped CLNGG is 3 × 3 × 6 mm^3. Both its 3 × 3 mm^2 end faces are AR-coated for the pump and laser wavelengths. To mitigate the thermal load, the crystal was wrapped with indium foil and tightly mounted in a copper holder with 14.0 °C water cooling. M_1 and M_2 are plano-concave mirrors both with radius-of-curvature of −100 mm. Plane-wedged mirrors with partial reflection at the laser wavelength are used as the output couplers (OCs). A Lyot filter is employed for the wavelength tuning. CM_1 and CM_2 are chirped mirrors with group delay dispersion (GDD) of −125 fs^2 per bounce. The SWCNT-SA fabricated by spin coating SWCNT/PMMA (polymethyl methacrylate, ~300 nm [17]) films onto a 1 mm thick quartz substrate was inserted under Brewster's angle in the second cavity waist. Its non-saturable loss around 2 µm is ~1%, associated with < 0.5% modulation depth and < 10 µJ/cm^2 saturation fluence. The SWCNT-SA can withstand incident peak fluence as high as 1 mJ/cm^2 even at shorter wavelengths [18]. The calculated beam radii on the SWCNT are 70 µm in the sagittal and 110 µm in the tangential plane.

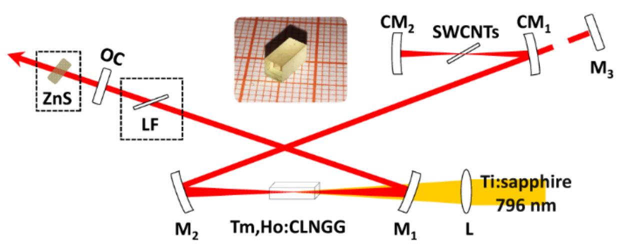

Fig. 1. Scheme of the tunable and mode-locked Tm,Ho:CLNGG laser. (L, lens; M: mirrors, CM, chirped mirrors; LF, Lyot filter; OC, output coupler).

With the X-folded cavity in Fig. 1, CW and wavelength tunable operation were firstly studied. The physical cavity length was about 1.5 m and the transmission of the OCs was 0.5%, 1.5%, 3%, and 5%. The crystal absorption under lasing conditions with 3% OC decreased from 73.3% to 68.3% with increasing the incident pump power, which was caused

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by the absorption bleaching under tight focusing. Figure 2(a) shows the corresponding output power for the different OCs. A maximum output power of 0.485 W was obtained with 3% OC, corresponding to a slope efficiency of 24.2%. The reduction of slope efficiency for 5% OC can be attributed to the stronger upconversion effect under the higher population of the upper laser level. As expected, the central wavelength presented a red shift from 2080 to 2096 nm when decreasing the OC transmission, which as a typical feature of quasi-three-level laser systems related to the stronger reabsorption effect of the larger ground state populations for lower OC transmission. As shown in Fig. 2(b), a wavelength tuning range of 205 nm, i.e., from 1924 to 2129 nm, was achieved with a Lyot filter (3.2 mm thick quartz plate with the optical axis at 60° to the surface) in the cavity, larger than the 174 nm obtained with the Tm,Ho:CNGG laser [8] under almost the same conditions. Such a broad tuning range lying outside the water vapor absorption region, as shown by the green line in Fig. 2(b), is a prerequisite for stable femtosecond pulse generation.

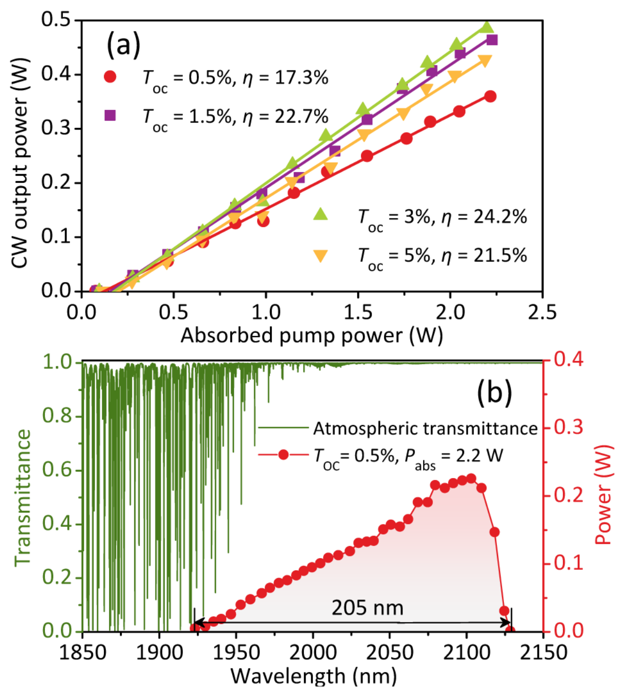

Fig. 2. CW input-output performance (a) and wavelength tunability (b) of the Tm,Ho:CLNGG laser. η is the slope efficiency. The green line in (b) is atmospheric transmittance at normal conditions for a path length of 3 m (HITRAN database, USA model, high latitude, summer, H = 0).

Mode-locking was studied by substituting M_3 with chirped mirrors (CM_1 and CM_2) and inserting the SWCNT-SA into the cavity. Taking into account the group velocity dispersion of the 6-mm thick CLNGG crystal (~-34 fs^2/mm at 2080 nm) and the 1-mm thick quartz substrate of the SA (~-118 fs^2/mm, placed at Brewster angle), the total round trip GDD amounted to −1080 fs^2 as shown in Fig. 3(a). By carefully adjusting the beam position on the SWCNT-SA and optimizing the separation between M_1 and M_2, stable, self-starting mode-locking was achieved with no multi-pulsing or satellites. With 3% OC, the threshold for transition from unstable Q-switching regime to self-starting mode-locking was at an absorbed pump power of ~1.35 W. Increasing the pump power to 2.2 W, an average output power of 123 mW was achieved at a repetition rate of 99.28 MHz, equivalent to a single pulse energy of 1.24 nJ. In this case, the average fluence on the SWCNT-SA was calculated to be ~170

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μJ/cm^2. The recorded laser spectrum and the intensity autocorrelation trace are shown in Fig. 3(a) and 3(b), respectively. The central wavelength was located at 2069 nm with a full width at half maximum (FWHM) of 46.3 nm, and the pulse duration was 98 fs by assuming sech^2-intensity profile, yielding a time-bandwidth product (TBP) of $\Delta\nu\tau$ = 0.318, very close to the Fourier-transform limit.

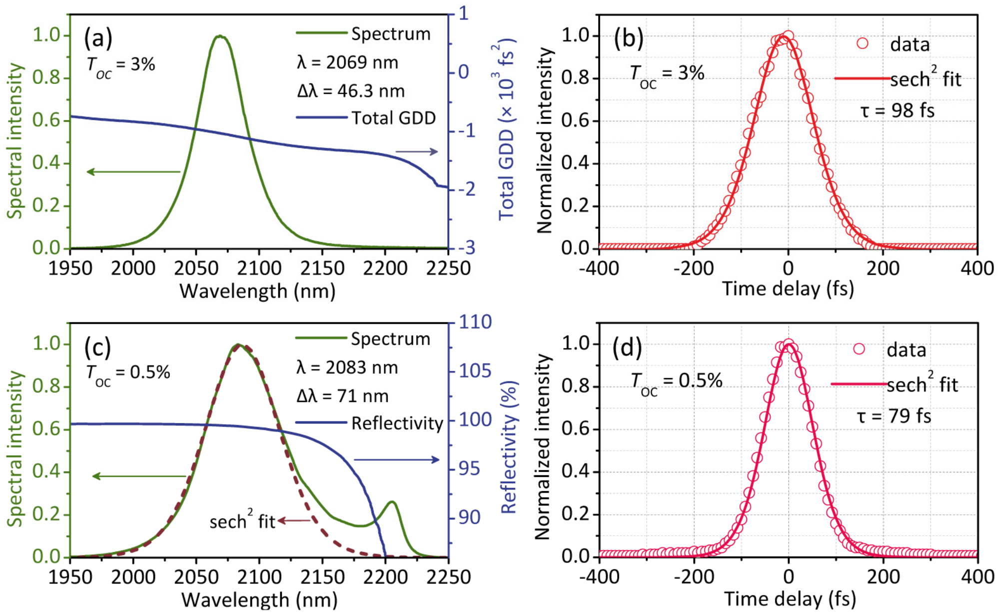

Fig. 3. (a), (c) Optical spectra and (b), (d) the corresponding noncollinear autocorrelation traces of the mode-locked Tm,Ho:CLNGG laser for OC transmission of T_{OC} = 3% and 0.5%, respectively. The blue line in (a) is the calculated total intracavity GDD. In (c) the dashed line is the sech^2 fit of the optical spectrum and the blue line is the reflectivity of the 0.5% OC.

One notable feature of this laser was that sub-100 fs pulse generation was achieved in the second mode-locking region by only adjusting the separation between M_1 and M_2 whereas in the first region the pulse durations exceeded 200 fs. This is attributed to the balance of the GDD and self-phase modulation (SPM) as a result of the tight mode focusing condition, i.e., soliton mode-locking [19]. However, pure Kerr-lens mode-locking of the Tm,Ho:CLNGG laser was impossible by substituting the SA with a 1-mm thick quartz plate, which indicated that the SWCNT-SA was essential for starting and stabilizing the mode-locking while SPM was the main factor enabling sub-100 fs pulse generation.

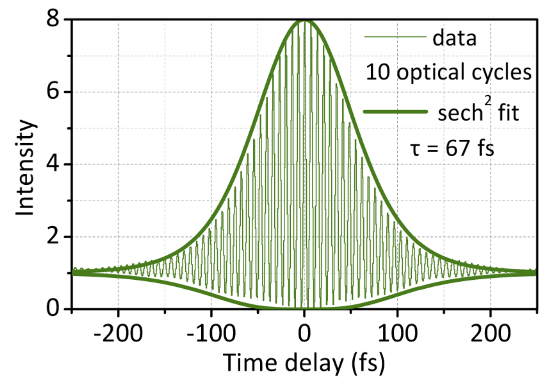

Fig. 4. Interferometric autocorrelation trace of the mode-locked laser after extra-cavity compression.

To further shorten the pulse duration, the 0.5% OC was employed. Similarly to the case with 3% OC, mode-locking was self-starting and stable for hours. The maximum output power in the mode-locking regime was 40 mW, corresponding to an average energy fluence of ~330 μJ/cm^2 on the SWCNT-SA. The optical spectrum centered at 2083 nm with a spectral FWHM of 71 nm is shown in Fig. 3(c). As can be seen, a sideband around 2205 nm is present in the right shoulder of the spectrum. It can be attributed to leakage since the OC transmission dramatically increases to 14% at this wavelength, i.e., nearly 30 times compared to 2083 nm as shown by the blue line in Fig. 3(c). According to the sech^2 fit of the optical spectrum, the spectral FWHM amounts to 69 nm. Figure 3(d) shows the corresponding noncollinear autocorrelation trace. Pulses with a FWHM of 79 fs were achieved, which gives a TBP of 0.377. The larger value compared to the case for 3% OC means that further shortening is possible by suitable dispersive elements.

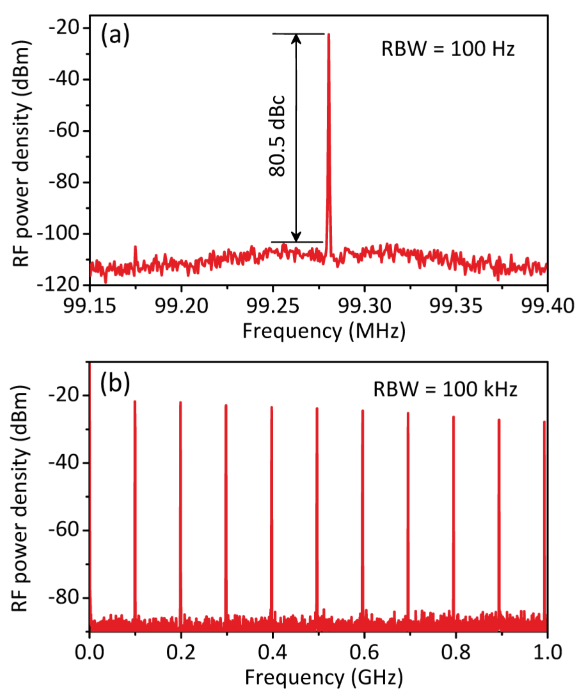

Fig. 5. RF spectra of the fundamental beat note (a) and a 1 GHz span (d) for the 10-optical-cycle pulses. RBW: resolution bandwidth.

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Thus a 3-mm-thick ZnS plate with GDD of + 462 fs^2 was used as an extracavity compression element. After passing the laser through the ZnS plate, the pulses were compressed to as short as 67 fs, leading to a TBP of 0.319. The interferometric autocorrelation trace is shown in Fig. 4 with the expected peak-to-background ratio of 8:1 [20]. An almost perfect fit was obtained with envelope profiles corresponding to an unchirped $sech^2$ pulse shape [20]. On the other hand, from the real-time fringe-resolved interferometric information we can confirm that the pulse duration is ~10 optical cycles, which to our knowledge are the shortest pulses ever reported with such CNGG-type crystals in the 2 μm spectral range. Single pulse operation was further confirmed by checking the intensity autocorrelation trace on a longer time scale of 50 ps. Apart from the attractive features of Tm,Ho:CLNGG, we attribute the generation of such short pulses with stable and clean $sech^2$ temporal profile to the relatively large nonlinear refractive index n_2 of this crystal (calculated to be 7.3×10^{-20} m^2/W around 2 μm, larger than that the 1.6×10^{-20} m^2/W value estimated for Tm,Ho:$KY(WO_4)_2$ [21] and the 3.3×10^{-20} m^2/W for Tm:Lu_2O_3 [22]). This is expected to enhance the SPM effect under tight mode focusing conditions leading to the soliton-like pulse shaping mechanism.

The radio frequency (RF) spectra were recorded to further characterize the stability of the mode-locking for the 10-optical-cycle pulses. As seen in Fig. 5(a) and 5(b), the high extinction ratio of 80.5 dBc above the noise level of the fundamental beat note recorded on a 250 kHz span with 100 Hz resolution bandwidth (RBW), indicates a stable steady-state mode-locking. The uniform harmonic beat notes shown in Fig. 5(b) present a further evidence for stable and clean CW mode-locking without any multi-pulse behavior [23].

3. Conclusion

In conclusion, we have experimentally demonstrated the first CW and mode-locked operation of a Tm,Ho:CLNGG laser. In the CW regime, the Tm,Ho:CLNGG laser produced 0.485 W output without any thermal roll-over. A broad wavelength tuning range of 205 nm from 1924 to 2129 nm was realized with a Lyot filter, which revealed the potential for femtosecond pulse generation. By employing a SWCNT-SA and dispersion compensation (GDD = −1080 fs^2), sub-100 fs (98-fs) pulses were generated at a central wavelength at 2069 nm, corresponding to 123 mW output power and 1.24 nJ single pulse energy. With a 0.5% OC, the pulses were further shortened to 67 fs, i.e., 10 optical cycles, after extracavity compression and the corresponding optical spectrum was located at 2083 nm with a $sech^2$-fitted FWHM of 69 nm, thus giving a TBP of 0.319, very close to the Fourier-transform limit. Generation of such short pulses is believed to be facilitated by the broad and smooth spectral gain profile, the long wave emission above 2 μm, and the large nonlinear refractive index of the Tm,Ho:CLNGG crystal which enhances the SPM effect. Future work will focus on further shortening the pulse duration by enhancing the SPM effect and optimizing the intracavity dispersion, in particular the 3rd order dispersion. Pure Kerr-lens mode-locking of such lasers based on CNGG-type crystals would be expected in the 2 μm spectral range due to the large nonlinear refractive index.

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