Single-walled carbon nanotube saturable absorber assisted high-power mode-locking of a Ti:sapphire laser

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Abstract: We report on passive mode-locking of a Ti:sapphire laser employing a single-walled carbon nanotube saturable absorber (SWCNT-SA) specially designed and fabricated for wavelengths near 800 nm. Mode-locked pulses as short as 62 fs were generated at a repetition rate of 99.4 MHz. We achieved output powers from the SWCNT-SA mode-locked laser as high as 600 mW with a slope efficiency of 26%. The characteristics of SWCNT-SA-assisted mode-locking were compared with those of Kerr-lens mode-locking without SWCNT-SA.

OCIS codes: (140.4050) Mode-locked lasers; (140.7090) Ultrafast lasers; (160.4236) Nanomaterials; (160.4330) Nonlinear optical materials.

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1. Introduction

Ti:sapphire lasers operating near 800 nm are widely exploited in laboratories as ultrafast light sources for a variety of applications. Owing to excellent 3rd-order nonlinear optical properties of the Ti:sapphire crystal, Kerr lens mode-locking (KLM) technique has been commonly used for passive mode-locking in Ti:sapphire lasers since the early 1990s [1]. Although KLM shows an almost instantaneous response and high power operation is possible in the absence of internal losses, typical for any other mode-locking device, it requires precise cavity alignment due to a narrow stability region where the Kerr lensing and mode-locked operation is stable. Hence, investigations on saturable absorbers, such as dielectrics doped with absorbing ions [2,3] and semiconductor structures [4–6], have been further conducted for convenient, stable, and self-starting solid-state laser mode-locking. Especially, semiconductor saturable absorber mirrors (SESAMs) became a standard mode-locking device for ultrashort pulse generation at different wavelengths because their absorption recovery time, saturation fluence and, only partly, spectral range, can be engineered by choosing suitable materials, growth parameters and device designs [7]. The combination of KLM and SESAM can ensure self-starting and stable operation of mode-locked Ti:sapphire lasers [5,8], but relatively high cost and complex manufacturing processes limited the wide use of such SESAMs.

Passive mode-locking techniques employing single-walled carbon nanotube saturable absorbers (SWCNT-SAs) have been extensively investigated as an alternative since the first demonstration of mode-locking in a fiber laser in 2004 [9]. SWCNT-SAs exhibit not only nonlinear optical response comparable to that of SESAMs but also low nonsaturable loss and high damage threshold [10,11]. Moreover, careful dispersion control of carbon nanotubes of different diameters in the suspension enables flexible SWCNT-SAs designs applicable for laser mode-locking in a wide spectral range. Recently, we demonstrated passive mode-locking of different bulk solid-state lasers employing SWCNT-SAs in the spectral range between 1 and 2 μm [12–15], where E_{11} and E_{22} electronic transitions of arc-discharge grown and E_{11} electronic transition of high pressure CO (HiPCO) conversion SWCNTs were utilized. From the currently available SWCNTs, only HiPCO SWCNTs show broad absorption around 800 nm. However, it is not a trivial task to achieve optimized absorption band and saturable absorption at this wavelength because the absorption band edge of semiconducting E_{22} transitions of HiPCO SWCNTs is approached in this spectral range and also the metallic E_{11} band is lying close to 800 nm [16]. Moreover, SWCNTs exhibit quite large two-photon absorption (TPA) near 800 nm [17]. Up to now, there is only one report on SWCNT-SA-

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based passive mode-locking of a Ti:sapphire laser [18]. In this report, output power of only 45 mW was obtained at quite a high pump power of 4 W and there was no intra-cavity dispersion compensation. This low power was a consequence of the high intra-cavity losses and damage limitation of the SWCNT-based saturable absorber used.

In the present work, we present passive mode-locking of a Ti:sapphire laser, generating 62 fs pulses by intra-cavity chirp compensation, with a transmission-type SWCNT-SA optimized for 800 nm. The characteristics of the mode-locked regime, such as threshold, stability, pulse durations and spectral bandwidths are compared with those of pure KLM, i. e. mode-locking without the SWCNT-SA.

2. Characteristics of SWCNT-SA

Purified SWCNTs from Unidym Inc. synthesized by HiPCO conversion technique were used as the saturable absorber material. After dispersing SWCNTs of 0.15 mg/ml in dichlorobenzene (DCB) via ultrasonic agitation for 2 hours, the dispersion was further centrifuged for accurate segregation. The well-dispersed SWCNTs/DCB solution was then mixed with PMMA solution, spin coated on a quartz window and baked at 170°C. The thickness of the SWCNT absorber layer was measured to be 400 nm.

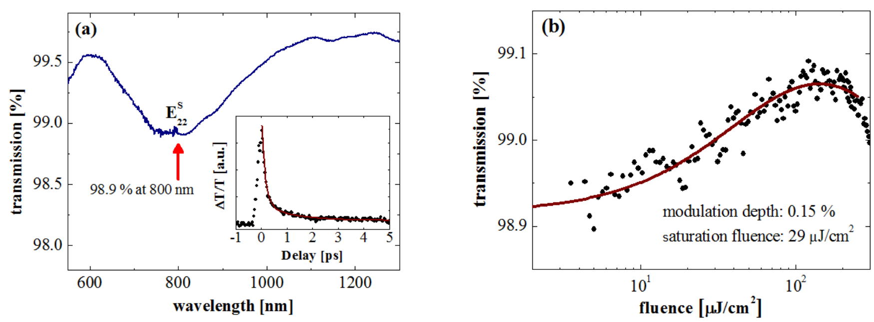

Fig. 1. Characteristics of transmission-type SWCNT-SA at 800 nm: (a) linear transmission spectrum and nonlinear dynamics (inset) and (b) nonlinear absorption.

The SWCNT-SA fabricated with optimized dispersion yields a well-defined broad absorption band, which corresponds to the E_{22} interband transition of SWCNTs. Figure 1(a) shows the linear transmission spectrum of the SWCNT-SA with absorption peak located at the gain window of Ti:sapphire. The linear transmission at 800 nm amounted to 98.9%. The nonlinear response of the SWCNT-SA was measured at 800 nm by non-collinear cross-polarized pump-probe spectroscopy. The pump-probe trace and a fit to the measured data are shown in the inset of Fig. 1, indicating a nearly instantaneous response of about 150 fs and a slow exponential decay of < 1.2 ps. These intraband and interband recovery times are comparable to the values measured for the E_{11} interband transition of HiPCO SWCNTs, which was previously utilized for Cr-doped bulk lasers such as Cr:forsterite and Cr:YAG lasers in our mode-locking experiments [13,14]. The nonlinear absorption behavior of the SWCNT-SA in terms of saturation fluence and modulation depth was characterized by nonlinear transmission measurements at 800 nm with a setup similar to that described in [19], but modified for transmission measurements. In contrast to previous SWCNT-SAs which were used for bulk laser mode-locking above 1 μm spectral range, a rollover was clearly seen in the transmission of the present sample at fluences exceeding 200 μJ/cm^2 for wavelengths near 800 nm, obviously caused by TPA (see Fig. 1(b)). However, as it will be seen, this had no noticeable effect on the stable mode-locking up to output powers of 600 mW. From the fit to the data we extracted an effective modulation depth of about 0.15% and a saturation

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fluence of about 29 μJ/cm^2. These parameters are suitable to achieve stable passive mode-locking without Q-switching instabilities [20].

3. Experimental setup

The schematic layout of the laser setup is shown in Fig. 2. A x-folded 5-mirror resonator was used for the laser experiment. The 4-mm-long Brewster-cut Ti:sapphire crystal was mounted in a copper block and a temperature of 14°C was maintained by water cooling. A continuous-wave (CW) OPSL (Coherent Inc., Verdi G5) operating at 532 nm was used as the pump source. Two dielectric concave mirrors with a radius of curvature (ROC) of −100 mm were arranged with a folding angle of 14.5° to compensate the astigmatic aberration. Two highly reflecting concave mirrors (M3 and M4) with the same ROC were used to form the additional intracavity waist where the SWCNT-SA was mounted under Brewster angle to minimize the cavity loss. For stable laser operation, the M1-M2 and M3-M4 separations were kept at about 11 and 15 cm, respectively. For this alignment, the measured beam radius at the second cavity waist was about 40 μm. To avoid possible damage of the SWCNT-SA, it was positioned out of the focus. An output coupler of 10% transmission was employed for extraction of sufficient output power. A SF10 prism pair with a separation of 16 cm provided negative dispersion. Mode-locked operation with SWCNT-SA was compared to pure KLM in the same configuration. For spectral tuning of the mode-locked laser, a knife edge between P1 and P2 was used in addition to slightly translating the prism P2.

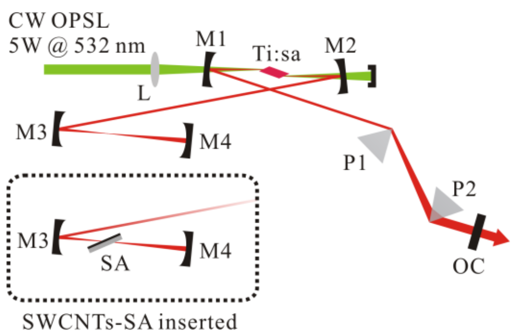

Fig. 2. Configuration of the Ti:sapphire laser mode-locked by KLM and SWCNT-SA (dashed box): L: focusing lens with f = 100 mm; Ti:sa: 4-mm-long Ti:sapphire crystal; M1-M4: HR-coated concave mirrors with a ROC = −100 mm; P_1 and P_2: SF10 prisms; OC: 10% output coupler; SA: SWCNT-SA.

4. Mode-locked operation

For characterization of mode-locked operation initiated by two different techniques, we kept the repetition rate and especially the separation of M3 and M4 nearly constant in the experimental setup. In the case of pure KLM, the power range in which the laser was stably mode-locked was much narrower than that with SWCNT-mode-locking. In addition, the operation was not self-starting and more sensitive to cavity alignment. When the SWCNT-SA was used, we were able to achieve stable mode-locking without critical alignment in a wider power range. In order to suppress KLM, we slightly changed the position of M2 so that mode-locking was not self-starting by KLM alone. Since the laser still showed a tendency to mode-lock, we call the regime where the SWCN-SA contributes to self-starting and stabilization SWCNT-SA-assisted mode-locking. In this regime, the mode-locked laser operated stable for many hours with a negligible change of output power. Figure 3 shows the output powers versus the incident pump powers for the SWCNT-SA-assisted mode-locking and the pure KLM without SWCNT-SA. A slope efficiency of ~26% was obtained in both cases.

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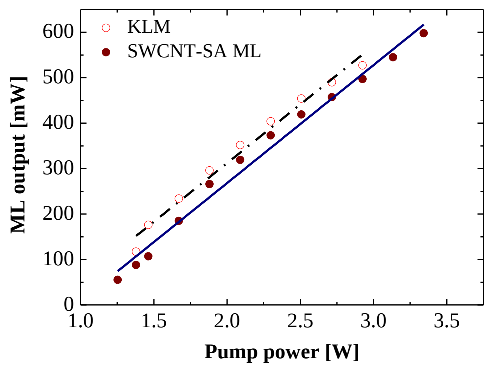

Fig. 3. Average output powers in the mode-locked (ML) regime versus incident pump powers of the KLM and the SWCNT-SA mode-locked Ti:sapphire laser with a linear fit to the data.

In case of the pure KLM, mode-locking started first at an output power of 117 mW with 1.38 W pumping, whereas the mode-locking threshold with the SWCNT-SA was lower, with 55 mW output power obtained at a pump power of 1.25 W. In the present laser setup, pure KLM delivered maximum output power of 527 mW while SWCNT-SA-assisted mode-locking produced higher output powers of 600 mW in a stable operation. Thus, although the SWCNT-SA introduced additional intracavity losses, we achieved lower mode-locking threshold and more stable operation with higher output power. We could not observe self-starting operation in the whole power range, but mode-locking was easily initiated by tapping of the end mirror or the SWCNT-SA.

The pulse durations in both mode-locking regimes near 800 nm were measured at the highest power level by the intensity autocorrelation technique using SHG in a 0.2 mm-thick type-I BBO. Shorter pulse duration and broader spectral bandwidth were obtained with mode-locking assisted by the SWCNT-SA. When a $sech^2$-shaped pulse is assumed, pulse durations of 70 and 62 fs and corresponding spectral bandwidths of 12.5 and 14.2 nm were achieved in pure KLM and SWCNT-SA-assisted mode-locking, respectively. The autocorrelation trace of SWCNT-SA mode-locked pulses is depicted in Fig. 4(a).

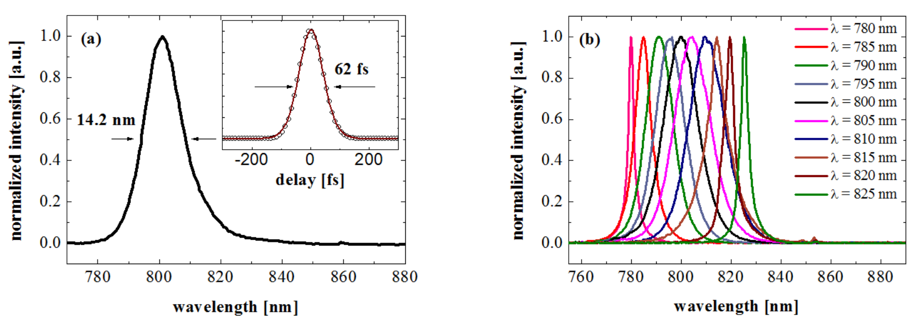

Fig. 4. SWCNT-SA mode-locked Ti:sapphire laser: (a) spectrum and autocorrelation trace with $sech^2$ pulse shape fit (inset) and (b) spectral tuning.

The time-bandwidth product of 0.41 indicates nearly Fourier transform-limited pulses. Figure 4(b) shows the spectral tunability of the SWCNT-SA mode-locked Ti:sapphire laser. A continuous spectral tuning over 45 nm from 780 to 825 nm was easily realized by moving the knife edge between the two prisms and the prism P2. In spite of the spectral narrowing

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approaching the edge of the dielectric mirror coating, no fluctuation of output power or instability of mode-locking were observed.

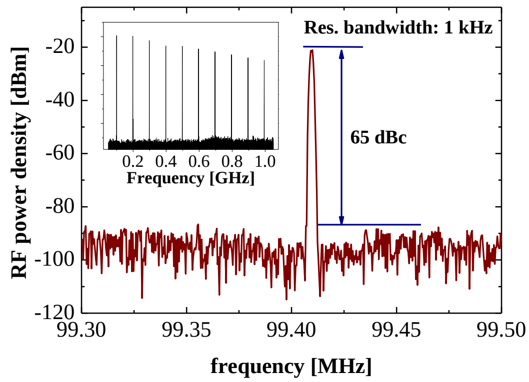

Fig. 5. RF spectrum of the fundamental beat note in 200 kHz and 1 GHz (inset) span.

Stable CW mode-locked operation without Q-switching modulation and multiple pulsing was verified by the radio-frequency (RF) spectrum measurement. Figure 5 shows the recorded RF spectrum at the fundamental beat note of 99.4 MHz in a 200 kHz span with a resolution bandwidth of 1 kHz. A high extinction ratio of 65 dBc was obtained. The inset depicts a wide-span RF measurement.

5. Conclusion

In conclusion, we fabricated transmission-type SWCNT-SAs with optimized SWCNT dispersion applicable near 800 nm and demonstrated mode-locking of a Ti:sapphire laser delivering output powers as high as 600 mW. Even though TPA is in principle present at high fluences near 800 nm, this SWCNT-SA with suitable recovery times and modulation depth showed excellent performance as a passive mode-locking element. We confirmed that, in contrast with the general belief, SWCNT-SA-assisted mode-locking enables shorter pulse durations and broader spectral bandwidths compared to pure KLM. Thus, low-cost and properly engineered SWCNT-SAs can be successfully used as an alternative to widespread SESAMs for ultrafast Ti:sapphire lasers.

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