Mode-locked 2 μm fiber laser with a multi-walled carbon nanotube as a saturable absorber

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Received October 7, 2014; accepted December 22, 2014; posted online February 24, 2015

We propose and demonstrate a passively mode-locked fiber laser operating at 1951.8 nm using a commercial thulium-doped fiber (TDF) laser, a homemade double-clad thulium-ytterbium co-doped fiber (TYDF) as the gain media, and a multi-walled carbon nanotube (MWCNT) based saturable absorber (SA). We prepare the MWCNT composite by mixing a homogeneous solution of MWCNTs with a diluted polyvinyl alcohol (PVA) polymer solution and then drying it at room temperature to form a film. The film is placed between two fiber connectors as a SA before it is integrated into a laser ring cavity. The cavity consists of a 2 m long TDF pumped by a 800 nm laser diode and a 15 m long homemade TYDF pumped by a 905 nm multimode laser diode. A stable mode-locking pulse with a repetition rate of 34.6 MHz and a pulse width of 10.79 ps is obtained when the 905 nm multimode pump power reaches 1.8–2.2 W, while the single-mode 800 nm pump power is fixed at 141.5 mW at all times. To the best of our knowledge, this is the first reported mode-locked fiber laser using a MWCNT-based SA.

OCIS codes: 060.3510, 320.7090. doi: 10.3788/COL201513.030602.

Light sources operating at 2 μ m lasers have been extensively investigated in recent years due to their many potential scientific applications, particularly in the fields of spectroscopy, medicine, remote sensing, and communications. This wavelength also happens to coincide with many absorption lines of several gas molecules, e.g., carbon dioxide (CO₂) and hydrogen bromide (HBr)^[1]. This creates the possibility of constructing various trace-gas sensors. The wavelength also has a strong water absorption characteristic, which might be useful in various biomedical applications. It was reported that a 2 μ m source outperformed 1 and $1.55 \ \mu m$ sources when used in dermatology and surgery as an optical scalpel $\left[\frac{2,3}{2}\right]$. The light source operating in the range of $1.8-2.0 \ \mu m$ can be realized using a thulium-doped fiber laser (TDFL)^[4]. Modelocking techniques can also be used for generating ultrashort pulse TDFLs, which can extend the applications of those sources.

Till now, mode-locked TDFLs are usually achieved using nonlinear polarization rotation $(NPR)^{[5]}$, semiconductor saturable absorber mirrors $(SESAMs)^{[6]}$, and single-walled carbon nanotubes $(SWCNTs)^{[7]}$. Although these approaches are well established, NPR-induced lasers tend to be environmentally unstable and do not provide self-starting pulsed operation. SESAM-based lasers have a limited operating band. Lasers produced using SWCNT saturable absorbers (SAs) are known to have an ultra-fast recovery time and a wide absorption bandwidth^[8,9]. A new member of the carbon nanotube (CNT) family, multiwalled CNTs (MWCNTs)^[10,11], have also gained much attention as an alternative to SWCNTs for nonlinear optics applications. They possess similar characteristics to the SWCNTs but have a lower production cost; they are 50%–80% cheaper than the SWCNT materials^[12]. Compared to SWCNTs, MWCNTs have greater mechanical strength and photon absorption per nanotube, and better thermal stability, due to their higher mass density^[13,14].

Recently, Lin *et al.*^[15] employed a MWCNT-based SA to generate mode-locking pulses in a Nd:YVO₄ laser. In this Letter, to the best of our knowledge, a mode-locked 2 μ m fiber laser is demonstrated for the first time. We use a simpler and lower-cost MWCNT-based SA in which a commercial thulium-doped fiber (TDF) and homemade double-clad thulium-ytterbium co-doped fiber (TYDF) are used as gain media. Both gain media are cascaded to produce an amplified spontaneous emission in the 1950 nm region, which requires a high gain to initiate the mode-locking process at 1951.8 nm. The SA is constructed by sandwiching a MWCNT-polyvinyl alcohol (PVA) film between two fiber connectors.

The SA was fabricated using a MWCNT powder with a diameter of 10–20 nm and length of 1–2 μ m as the raw material. The MWCNT was functionalized with a sodium dodecyl sulfate (SDS), to make it soluble in water. We mixed 4 g of SDS with 400 ml of deionized water before 250 mg of MWCNT was added to the solution. The

mixture was sonicated for 60 min at 50 W to obtain a homogeneous dispersion of MWCNTs that was then centrifuged at 1000 rpm to remove any large particles of undispersed MWCNTs. This dispersed suspension is expected to be stable for a few weeks. The dispersed MWCNT suspension was mixed with a PVA solution at a ratio of one to four to obtain a MWCNTs-PVA composite. In our experiment, the PVA solution was prepared by dissolving 1 g of PVA ($Mw = 89 \times 103$ g/mol) in 120 ml of deionized water. The composite was sonificated for more than one hour to realize a homogeneous MWCNTs-PVA precursor. Then, it was cast onto a glass petri dish and left to dry at room temperature for about one week to produce a film with a thickness of around 50 μ m. The SA was fabricated by sandwiching a small clipping of the film between two FC/PC fiber connectors. An index-matching gel was applied to the fiber ends of the connectors before matching, to reduce the insertion loss. The insertion loss of the SA was approximately 3.3 dB at 1900 nm.

Figure 1 shows the Raman spectrum, which is obtained by exciting the MWCNT-PVA film using a 352 nm laser. It shows well-defined G (1585 cm⁻¹) and D (1433 cm⁻¹) bands in Fig. <u>2</u>, as are normally observed in the spectra of graphite or graphene. The G-band is associated with the in-plane tangential stretching of the carbon-carbon bonds in the graphene sheet. The D-band is affiliated with the hybridized vibrational mode of the CNT edges, which indicates the presence of some disorder in its structure. The existence of a prominent D-band in the Raman spectrum shows that the CNTs are the multi-walled type with a multi-layer configuration and a disordered structure. The D-band, which is less intense than the G-peak, is reported to be upshifted to 1275 cm^{-1} in the MWCNT-PMMA (poly(methyl methacrylate) composite. In this work, we observed a downshift in the D-band for the MWCNT-PVA polymer composites. The absence of the radial breathing mode at the $\sim 250 \text{ cm}^{-1}$ band in the Raman spectrum confirms the multi-walled type of the CNT used in our experiment. This is due to the diameter of the wrapped CNT, which either consists of too many walls or has an inner wall diameter that is too



Fig. 1. The Raman spectrum obtained from the MWCNT-PVA film.



Fig. 2. The linear transmission curve of the MWCNT-PVA film, which shows the saturable absorption and non-saturable absorption values of around 4% and 45%, respectively.

big. Furthermore, other prominent features, like the D+G band (2900 cm⁻¹) and a small peak at 845 cm⁻¹, can also be observed in the Raman spectrum of Fig. <u>1</u>. A linear transmission curve of the MWCNT-PVA film is also investigated, and the result is illustrated in Fig. <u>2</u>. The curve indicates that the film has a saturable absorption (modulation depth) of around 4%, and a non-saturable absorption of around 45%.

Figure 3 shows the experimental setup of the proposed laser, using the fabricated MWCNT-PVA film as a SA. A 2 m long TDF pumped by an 800 nm laser diode via an 800/2000 nm wavelength division multiplexer and 15 m long homemade TYDF are used as gain media. The TYDF is pumped by a 905 nm multimode laser diode via a multimode combiner. The TDF has core and cladding diameters of 9 and 125 μ m, respectively, a loss of less than 0.2 dB/km at 1900 nm, and a Tm ion absorption of 27 dB/m at 793 nm. The homemade TYDF was drawn from a lithium-aluminum-germanosilicate core glass optical preform. The preform was fabricated via the modified chemical vapor deposition (MCVD) process, followed by a solution doping technique. The preform consists of Al₂O₃, Y_2O_3 , Tm_2O_3 , and Yb_2O_3 dopants with average weight percentage of 5.5, 3.3, 0.7, and 4.0, respectively. It has



Fig. 3. Experimental setup of the proposed mode-locked 2 μ m fiber laser, using a hybrid gain media in conjunction with a MWCNT-PVA film SA. The inset shows the microscopic cross-sectional view of an octagonal-shaped, double-clad TYDF.

an octagonal shaped double-clad structure, as shown in the inset of Fig. 3, to improve the pump absorption efficiency. The doping levels of the Tm^{3+} and Yb^{3+} ions of the fabricated TYDF are measured to be around 4.85×10^{19} and 27.3×10^{19} ions/cc, respectively, using an electron probe micro-analyzer. The NA and core diameter of the fabricated TYDF are measured to be 0.23 and 5.96 μ m, respectively. The splicing loss between this fiber and other fibers in the cavity is less than 0.1 dB. The output laser is tapped via a 10 dB coupler, which keeps 90% of the light oscillating in the ring cavity. The cavity length is measured to be approximately 23.5 m. A polarization controller (PC) is utilized to tune the polarization state of the light in the laser cavity. An optical spectrum analyzer (OSA) with a spectral resolution of 0.05 nm is used for the spectral analysis of the mode-locked laser, and an oscilloscope is used to analyze the output pulse train of the mode-locking operation via a photo-detector. It is worth noting that we intentionally cascaded the TDF with TYDF in the laser cavity to obtain a laser operation at a longer wavelength in the 1950 nm region. The use of the cascaded gain media also reduces the threshold pump power for the lasing, as well as increases the gain in the 1950 nm region. Thus, it allows the mode-locking pulse to be generated in the cavity.

The output spectrum of the mode-locked laser was first monitored by an OSA that has a resolution that is limited to 0.05 nm. Figure <u>4</u> shows the optical spectrum when the 800 and 905 nm pump powers were fixed at 141.5 mW and 1.8 W, respectively. The laser operates at the central wavelength of 1951.8 nm, which is in the relatively longer wavelength region due to the use of the TYDF. The TYDF provides a higher gain at a longer wavelength, due to the transfer of energy from ytterbium to thulium ions. The full width at half-maximum of the output spectra is obtained at ~1.3 nm, which is slightly broadened due to the self-phase modulation effect in the laser cavity.

A stable mode-locking of the laser was observed as the 905 nm multimode pump powers were varied within the range of 1.8–2.2 W, and when the single-mode 800 nm pump power was fixed at 141.5 mW. At the beginning, the PC was slightly adjusted to initiate the mode-locking.



Fig. 5. Oscilloscope trace of a typical pulse train from the modelocked laser.

Figure <u>5</u> illustrates a typical pulse train output from the mode-locked laser when the 800 and 905 nm pump powers were fixed at 141.5 mW and 2.2 W, respectively. As shown in Fig. <u>5</u>, the peak to peak pulse duration for the laser is measured at 28.9 ns, which corresponds to a repetition rate of 34.6 MHz. The repetition rate was constant even when the multimode pump power was increased from 1.8 to 2.2 W. In order to show that the mode-locking operation was made possible by the MWCNT SA, the same laser configuration was also tested without the MWCNT-PVA film. But, no pulse train was observed in such a configuration at any point of the PC.

Figure <u>6</u> shows a plot of a typical output second harmonic generation autocorrelation trace superimposed on the sech² fit. Assuming a sech² profile, the pulse duration is estimated to be around 10.79 ps. The time-bandwidth product of the output pulses is ~11.05, with a major deviation from the value of 0.315 expected for the transform-limited sech² pulses. This shows that the pulse is highly chirped by the dispersion effect in the cavity, limiting the minimum achievable pulse width. Another limitation of shorter pulses could be due to the spectral filtering effects introduced by the Thulium gain medium.



Fig. 4. Optical spectrum of the mode-locked laser.



Fig. 6. Autocorrelation trace, with a sech² fit.



Fig. 7. RF spectrum of the mode-locked fiber laser.

It is also found that the average output power of the proposed mode-locked laser is linearly related to the pump power. The output power increases from 0.31 to 0.59 mW as the 905 nm pump power rises from 1.8 to 2.2 W, while the 800 nm pump power is fixed at 141.5 mW. At the maximum pump power, the pulse energy is calculated to be around 17.0 pJ. The radio frequency (RF) spectrum was also investigated, and the result is as depicted in Fig. 7. The repetition rate of the laser is 34.6 MHz, which corresponds to a fourth-order harmonic pulse with an approximately 23.5 m long cavity. A signal-to-noise ratio of about 20–25 dB is observed, highlighting the moderate amplitude fluctuation of the laser.

In conclusion, we demonstrate a passively mode-locked fiber laser operating at 1951.8 nm using a MWCNT SA. The laser uses a 2 m long TDF, which is pumped by an 800 nm laser diode and a 15 m long homemade TYDF pumped by a 905 nm multimode laser diode. The SA film is made from the MWCNT precursor, which is prepared by mixing the MWCNT homogeneous composite with a diluted PVA polymer solution. A small portion of the resulting film is sandwiched between two ferrules to form a SA, which is then integrated into the ring laser cavity. The

laser output generates a stable mode-locking pulse with a repetition rate of 34.6 MHz and a pulse width of 10.79 ps.

This work was supported by the University of Malaya Research Grant (UMRG) (Nos. RP008C-13AET and RU002/2013).

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