Carbon nanotube films for ultrafast broadband technology

Samuli Kivistö,[1,*] Tommi Hakulinen,[1] Antti Kaskela,[2] Brad Aitchison,[3] David P. Brown,[3] Albert G. Nasibulin,[2] Esko I. Kauppinen,[2] Antti Härkönen,[1] and Oleg G. Okhotnikov[1]

[1]*Optoelectronics Research Centre, Tampere University of Technology, P.O. Box 692, FIN-33101 Tampere, Finland*
[2]*NanoMaterials Group, Department of Applied Physics and Center for New Materials, Helsinki University of Technology, P.O.Box 5100, FIN-02015 TKK, Finland*
[3]*Canatu Ltd., Tekniikantie 21, FI-02150 Espoo, Finland*
[*]*Corresponding author: samuli.kivisto@tut.fi*

Abstract: Mode-locked sub-picosecond operation of Yb-, Er- and Tm:Ho-doped fiber lasers operating at 1.05 µm, 1.56 µm and 1.99 µm, respectively, is demonstrated using the same sample carbon nanotube-based saturable absorber mirror. A mesh of single-walled carbon nanotubes was deposited on an Ag-mirror using a one-step dry-transfer contact press method to combine broadband saturable absorption and high reflectance properties. The novel fabrication method of the polymer-free absorber and device parameters determined using nonlinear reflectivity measurement are described in detail. To our knowledge the observed operation bandwidth of ~1 µm is the broadest reported to date for a single carbon nanotube-based saturable absorber.

OCIS codes: (060.3510) Lasers, fiber; (140.4050) Mode-locked lasers.

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1. Introduction

Ultrafast passively mode-locked lasers are of unprecedented interest for a large variety of applications which has led to a recent search for novel, low-cost approaches. Passive mode-locking can be initiated by nonlinear polarization evolution, the Kerr-lens method or saturable absorption [1-3]. Saturable absorption can be generated by e.g. semiconductor saturable absorber mirrors (SESAMs) or various dyes [3,4]. Recently, new types of saturable absorbers based on single-walled carbon nanotubes (SWCNTs) have been used in fiber and solid-state lasers to initiate mode-locking [5-7]. The CNT-based devices benefit from a fairly simple manufacturing process, offer short absorption recovery times (<1 ps) and reasonable modulation depths [7,8]. Different SWCNT-absorbers have been reported to mode-lock lasers at wavelength regimes of 1 μm [6], 1.55 μm [5] and 1.9 μm [9]. However, none of the so far demonstrated absorbers have been shown to operate at all of these wavelengths. The operational parameters of a CNT-absorber can be tailored during the fabrication process since its absorption characteristics are determined by the diameter of the nanotubes and details of the structure [10].

CNT-based absorbers can be fabricated to operate both in transmission or reflection, which allows the use of either ring or linear cavity configuration [11]. Typically, CNTs have been embedded into polymer or liquid [7-9]. The CNTs, together with the polymer-based material, have then been applied on the surface of an optical fiber or a quartz substrate and used in transmission to mode-lock a ring-cavity laser [9,12,13]. Recently, a new method for placing SWCNTs onto a polymer film by a single-step dry-transfer thermo-compression method has been demonstrated, which is more practical compared with the conventional wet deposition methods [14,15]. Polymer is undesirable as an intracavity laser material due to its relatively low damage threshold and high optical absorption. It should be avoided, especially for high power and mid-IR applications. Therefore a new method to create CNT-based absorbers that does not employ polymer is very desirable.

In this paper, we report the use of a novel polymer-free carbon nanotube-based saturable absorber produced using a simple dry-transfer contact press method onto a highly reflective Ag-mirror. The same SWCNT-absorber mirror, used in a compact linear cavity configuration, is shown to provide self-starting sub-picosecond mode-locked operation of Yb-, Er- and Tm:Ho-doped fiber lasers at wavelengths of 1.05 μm, 1.56 μm and 1.99 μm, respectively. To our knowledge, this is the broadest operation bandwidth that has been reported to date for a single carbon nanotube-based saturable absorber.

2. Carbon nanotube production

SWCNTs were synthesized by thermal decomposition of ferrocene vapor in a carbon monoxide atmosphere [15]. Briefly, the Carbon Nanotube Reactor consisted of a saturator, a water-cooled injector probe, and a heated growth chamber. A 300 cm^3/min flow of CO was passed through the saturator filled with a mixture of silicon dioxide and ferrocene powders. This provided the conditions for the CO ferrocene vapor saturation of 0.8 Pa at room temperature. The growth chamber was an alumina tube inserted inside a tube furnace. The flow containing ferrocene vapor was then introduced directly into the high temperature zone of the growth chamber through the water-cooled probe. An additional CO flow (100 cm^3/min) was introduced outside the injector probe. The Ferrocene thermally decomposed in the high temperature gradient formed between the injector probe and the ambient reactor temperature. The decomposition leads to supersaturation conditions that result in iron nanoparticle formation. Iron nanoparticles are the catalysts from which the CNTs grow.

The reactor temperature was maintained at ~1100 °C. Catalytic CO disproportionation reactions resulted in the synthesis of single-walled CNTs. The product was collected downstream of the reactor by filtering the flow through a nitrocellulose membrane filter. The thickness and therefore optical transparency of the collected SWCNT-films can be accurately controlled by varying the collection time [14].

3. Absorber design and fabrication

A Saturable Absorber Incorporating NanoTubes (SAINT) was prepared by a room temperature dry-transfer process previously applied to SWCNT-films on polyethylene substrates [14]. A piece of a nitrocellulose membrane filter with a SWCNT-film was placed on a protected Ag-mirror with the SWCNT-film upside down. Then, the mirror and the filter were pressed together at a pressure of 1000 Pa. After the pressing procedure, the membrane filter was peeled off and the SWCNT-film was strongly adhered to the mirror surface. It is worth noting that the SWCNT-film was utilized as-deposited and no purification or dispersion steps were required. When compared to standard wet deposition methods, which may require several time-consuming stages, such as purification, dispersion and filtering, the approach of the SWCNT-film preparation demonstrated here is simple and easily scalable [16].

4. CNT-absorber mirror characterization

The SAINT-film was characterized by scanning and transmission electron microscopy, and optical absorption measurements. SWCNT-network morphology of the SAINT is depicted in the Fig. 1. SEM-imaging reveals up to 20 nm-wide bundles and iron nanoparticles which give large back-scattered electron yield and large contrast in the images. SEM-imaging of the SWCNT-network was performed on the nitrocellulose membrane filter prior to transfer. A

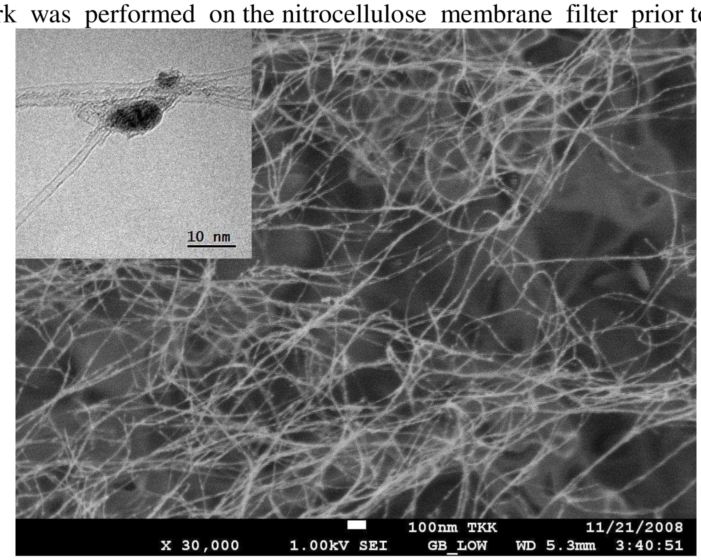

Fig. 1. SEM micrograph of the SWCNT-film. The inset shows high-resolution TEM-image of the sample. The tube diameter ranges from 1.2 nm to 1.8 nm.

sample of SWCNT-film was also transferred to a carbon coated TEM-grid for Carbon Nanotube diameter estimation. A high resolution TEM-image, shown as an inset to Fig. 1, reveals the tube diameter ranging from 1.2 nm to 1.8 nm.

Optical transmission spectroscopy was performed on a test sample using a dual-beam spectrophotometer. The test sample was prepared for this measurement by transferring

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another piece of the SWCNT-film to a PET-substrate using a dry-transfer process [15]. A blank PET-substrate was used in the reference beam. Figure 2(a) depicts the optical transmission spectrum of SWCNT-film with a thickness of ~80 nm. As can be seen, the absorption bands extend an ultra-broad wavelength range. The strongest absorption sub-bands of the SWCNT-film are located at 800 nm and 1400 nm. Optical uniformity of the SWCNT-film was estimated by measuring optical transmission over the SWCNT-film on PET-substrate at seven randomly located spots. Spectrophotometer beam diameter was limited to 1 mm. 1% standard deviation of optical transmission was observed at the wavelength of 550 nm, suggesting good optical uniformity of the SWCNT-film. The wavelength of 550 nm is typically used to characterize optical transmission of SWCNT-films [14].

Prior to use in laser experiments, the absorption saturation of the SAINT-mirror was determined from the dependence of the reflectivity on the incident pulse fluence for two wavelengths, 1.05 µm and 1.56 µm. The measurements were carried out using short pulse Yb- and Er-fiber systems. Figure 2(b) shows the nonlinear reflectivity at the wavelength of 1.56 µm. Non-bleachable loss (α_0), modulation depth (ΔR), and saturation fluence (F_{sat}) were determined to be 3.96%, 0.94% and 320 µJ/cm^2, respectively. At the wavelength of 1.05 µm the corresponding parameters were measured to be 18.3%, 1.86% and 1000 µJ/cm^2. We note that the Ag-mirror increases losses of the absorber by an additional 1-2%.

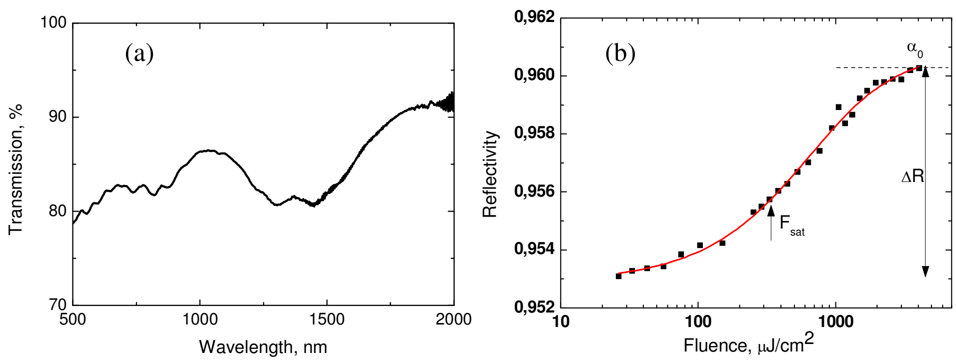

Fig. 2. (a) Optical transmission spectrum of the SWCNT-film. Ripples in absorption near 2000 nm wavelengths are caused by strong absorption of the PET-substrate. (b) Nonlinear reflectivity of the absorber measured at 1560 nm.

5. Mode-locked fiber laser experiments at 1.05 µm, 1.56 µm and 1.99 µm wavelengths

The same SWCNT absorber mirror was used as a mode-locking element in three lasers using Yb, Er and Tm:Ho doped fiber gain media with lengths of 1 m, 2 m, 1.5 m and operation wavelengths of 1 µm, 1.56 µm and 2 µm, respectively. The lasers had a linear cavity (see Fig. 3) that consisted of a pump laser, a wavelength selective fiber pump coupler, an ~10 % fiber output coupler, a highly reflecting dielectric end mirror and the SWCNT absorber mirror. A polarization controller was used in the cavity to optimize the laser performance. The Er- and Tm:Ho-doped fiber lasers operated in the soliton regime without dispersion compensation, while the Yb-doped fiber laser required a grating pair to compensate for the normal dispersion of an optical fiber. The Er- and Yb-lasers were pumped by a fiber-coupled 976-nm laser diode. The Tm:Ho laser was pumped by an Er-fiber laser emitting at 1.56 µm. The CNT absorber mirror, assembled with fiber butt-coupling, was typically oversaturated by the intracavity fluence of the laser.

The mode-locked Yb-fiber laser was operated at the central wavelength of 1050 nm with a

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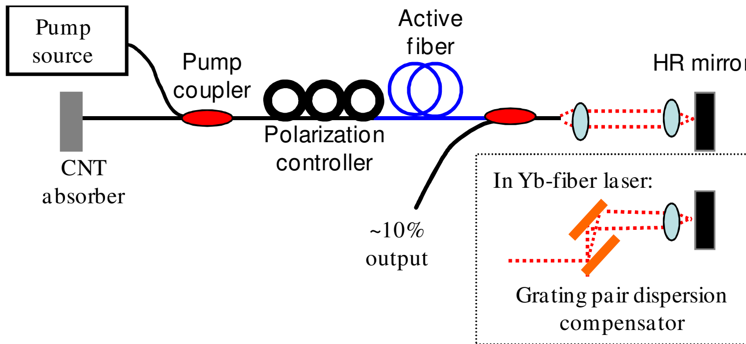

Fig. 3. Schematic of the experimental fiber laser setup used in the study. Active fibers acting as gain medium were doped with Yb, Er or Tm:Ho rare-earth materials. Yb-fiber laser requires the grating pair to achieve the soliton pulse regime.

spectral width of 2.7 nm, as seen in Fig. 4(a). Figure 4(b) shows the recorded autocorrelation trace with a $sech^2$-fitting revealing a pulse duration of 0.67 ps, resulting in a time-bandwidth-product of 0.48. The chirp could be expected from the strong dispersion management within the cavity and some additional chirp acquired in the output pigtail with a length of ~1.5 m. A photograph of a typical oscilloscope trace of the mode-locked pulse train is shown as an inset to Fig. 4(b). The average output power and threshold power were ~10 mW and ~150 mW, respectively. Once the laser operated in the mode-locked regime, the pump power could be reduced down to ~50 mW, revealing the hysteresis behavior typical for the soliton regime. With pump powers exceeding ~170 mW, multiple pulse operation expected for soliton fiber lasers was observed. The laser output characteristics are summarized in Table 1.

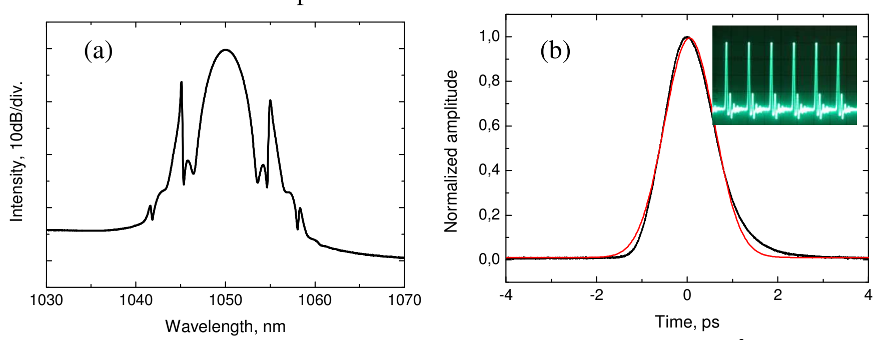

Fig. 4. (a) Pulse spectrum of the Yb-fiber laser. (b) Autocorrelation trace and $sech^2$-fit giving a pulse width of ~0.7 ps. The inset shows a typical oscilloscope picture of the generated pulse train.

The Erbium-doped fiber laser produced 0.44 ps pulses at a wavelength of 1563 nm, as seen in Fig. 5(a). The spectral width was measured to be 6.0 nm, giving a time-bandwidth product of 0.32 that corresponds to transform-limited pulses. The mode-locked operation was environmentally stable and self-starting with a low threshold pump power of ~30 mW.

The same SWCNT-absorber was also capable of initiating mode-locking in the thulium-holmium co-doped fiber laser. The polymer-free CNT-layer on a highly reflective Ag-mirror enabled the building of a robust and compact linear cavity laser with significantly improved output characteristics compared to the recently reported ring-cavity Tm-laser that had obvious limitations in output power, stability and absorber damage threshold [9]. The all-fiber laser delivered ~1.0 ps pulses with a central wavelength of 1991 nm, as shown in Fig. 5(b). The laser output parameters are summarized in Table 1. To our knowledge, this is the longest operation wavelength for a fiber laser mode-locked by a CNT-absorber. The extension in operation wavelength from ~1.9 μm [9] to ~2 μm is attributed to the use of a Tm:Ho co-doped fiber instead of a Tm-doped fiber with gain typically peaking at ~1.9 μm.

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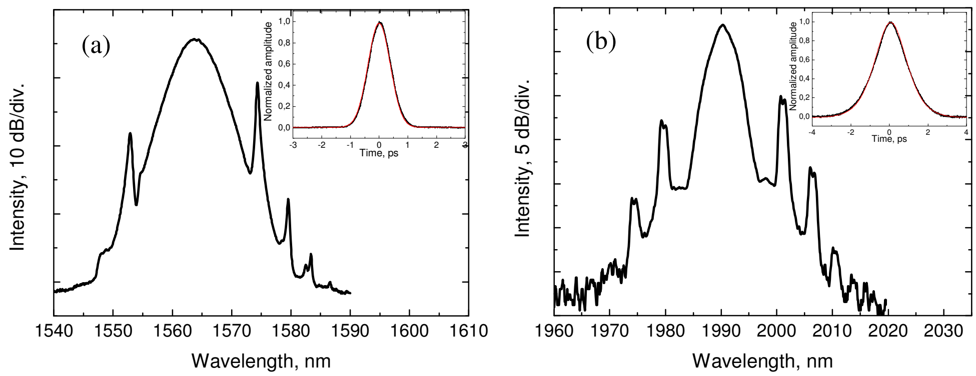

Fig. 5. Optical spectra of the Er- (a) and Tm:Ho-doped (b) fiber lasers. The corresponding autocorrelation traces with Sech^2-fittings are shown as insets. The pulse widths for the Er- and Tm:Ho-doped fiber lasers are 0.44 ps and 1.0 ps, respectively. The 3-dB spectral widths are 6.0 and 5.0 nm.

Table 1. Summary of the laser output characteristics.

	Threshold pump power, mW	Repetition rate, Mhz	Pulse width, ps	Time-bandwidth product	Average output power, mW
Yb	150	40	0.67	0.48	10
Er	30	15	0.44	0.32	10
Tm:Ho	150	41	1.0	0.37	15

All the mode-locking measurements were carried out in normal laboratory conditions at room temperature. The lasers were operated continuously for several hours at a time during the characterization. When the lasers were restarted after a few days, the mode-locked pulse train was immediately built up. No aging effects or degradation of the CNT-absorber at the used powers were observed during the measurements.

6. Conclusion

We have fabricated a novel saturable absorber mirror with single-walled carbon nanotubes transferred to an Ag-mirror using a simple contact press method. When compared to standard wet deposition methods, which require several processing stages, the presented approach for the SWCNT-film preparation is simple and scalable. Avoiding polymers as a host material, which could degrade the laser performance and increase absorber loss at wavelengths above 2 µm, allows for higher laser efficiency and power scalability.

Yb-, Er- and Tm:Ho-doped fiber lasers mode-locked using the same SAINT-mirror delivered sub-picosecond pulses at the wavelengths of 1.05 µm, 1.56 µm and 1.99 µm, respectively, thus demonstrating the ultra-broadband performance of the easily produced device. To our knowledge, this is the largest operation range reported to date for a SAINT-mirror. The use of the novel CNT-mirror technology offers an attractive opportunity for self-starting passive mode-locking of fiber lasers operating over a very broad spectral range with potential for further scaling to the mid-infrared spectral region and higher powers.

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