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Nonlinear absorption of SWNT film and its effects to the operation state of pulsed fiber laser

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Abstract: We study a single-wall carbon nanotube (SWNT) Polyvinyl alcohol (PVA) composite as a saturable absorber (SA) for pulse generation in Yb-doped fiber lasers. The saturable absorption and optical limiting (OL) characteristics of the SWNT device are investigated. By combing these two nonlinear effects, we find out for the first time, to the best of our knowledge, that mode-locking can be obtained in the dissipative soliton regime at low pumping followed by Q-switching at high pumping, which is quite different from conventional pulse dynamic evolutions. The Q-switched state operating at higher pump powers is due to the OL effect. The inverted operating fiber laser can be applied in various potential applications such as versatile material processing, optical communication and radar system etc.

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1. Introduction

Passively mode-locked and Q-switched fiber lasers have wide applications in materials processing, biomedicine, telecommunications, and radar system. Various kinds of saturable absorbers (SAs) have been utilized to achieve mode-locking in fiber lasers such as semiconductor saturable absorber mirrors (SESAM) [1–4], single-wall carbon nanotube (SWNT) [5–12], graphene [13,14], graphene oxide (GO) [15–17], and topological insulator (TI) [18–20]. However, SESAM has a low damage threshold, a complex semiconductor structure, a narrow turning range, and a limited recovery time [3]. Promising candidates are SWNTs, graphene, graphene oxide, and TI. In particular, SWNTs as excellent carbon materials have attracted considerable attention due to their broadband operation range, low cost, low saturation power, environmental robustness and short ultrafast recovery times (~1 ps) in the near-infrared region [6–10].

Generally, SAs can be used as mode lockers or Q switchers in a fiber laser under different conditions. Yb-doped mode-locked fiber lasers based on carbon nanotubes SA were demonstrated in several works [12,21,22]. Recently, passively Q-switched fiber laser has also been studied based on carbon materials and TIs [19,23–26]. B. Dong et al. reported a tunable passively Q-switched fiber laser with different cavity configurations [24]. J. Koo et al. reported a Q-switched and an active mode-locked fiber laser based on SWNT SAs. However, most of the reported SWNT SA based fiber lasers focused on either the mode-locking operation or the Q-switching operation. What is the pulse evolution of the fiber lasers enabled by carbon materials? Is it identical with the conventional fiber laser based on nonlinear polarization rotation (NPR) technique and other techniques? Several works have been published about this issue: for example, the suppression of Q-switched mode locking utilizing the inverse saturable absorption (ISA) was theoretically demonstrated previously [27]. In 2005, the ISA (i.e., optical limiting (OL) effect) for self-stabilizing passively mode-locked lasers by using SESAM was reported [28]. Vivien et al. gave an overview of OL in carbon nanotubes [29], which indicates the possibility of utilizing OL effect of nanotubes to improve the output performances of their mode-locked lasers.

In this paper, we study saturable absorption and OL effects of SWNT/PVA films. Then we incorporate the film in our Yb-doped fiber laser to adjust the output performance: The proposed fiber laser can operate in passively mode-locked states at low pump power and Q-switched states at high pump power. That is different from the conventional operating evolution in passively mode-locked fiber lasers. We attribute this inversed operating state to the OL effect of the SWNT/PVA SA. Dissipative solitons with a center wavelength of 1060 nm can be obtained in mode-locked states at a low pump power (typically from 47 mW to 80 mW). When the pump power increases from 80 mW to 120 mW, the proposed fiber laser could work in Q-switched states that the repetition rate increases from 30 kHz to 50 kHz and the pulse duration decreases from 2.7 μ s to 1 μ s. To the best of our knowledge, it's the first time to experimentally demonstrate that the mode-locked states firstly and then Q-switched states in the same fiber laser. The inversed operation mode can be applied in versatile material processing, optical communication and Radar system etc.

2. The SWNT SA fabrication and experimental setups

The SWNTs are grown by the electric arc discharge technique. The mean diameter of the SWNTs of 1.5 nm is selected to provide better saturable absorption for mode-locking at 1 μ m in our experiment. The vertical evaporate methods are utilized to preparing the SWNT/ polyvinyl alcohol (PVA) films. Since the SWNT/PVA film is formed in a cell. The film from the wall of the cell is used as SA. We cut the films into small pieces in the experiments.

Figure 1(a) shows the Raman spectrum of SWNT/PVA film. Different parameters such as diameter, electrical characteristic, direction, and chirality can be derived from the main firstand second-order Raman modes of the Raman spectrum [30]. Radial-breathing-mode (RBM) band (100-400 cm⁻¹) is induced from radial vibration of the carbon tube in phase, which can also be utilized to estimate the diameter of the tube. D band (1300-1400 cm⁻¹) is related with the breathing motions of the sp² carbon atoms in a ring. It reflects the defects on the nanotube surface. While G-band (1500-1600 cm⁻¹), usually consisting of two sub-bands, is related with the axial and circumferential in-plane vibrations. The average diameter of the SWNTs (*ds*, nm) can be estimated from the frequency (ω_{RBM} , cm⁻¹) of the RBM by the Eq. $\omega_{\text{RBM}} = A/ds + B$, where A and B are empirically found to be 223.5 nm·cm⁻¹ and 12.5 cm⁻¹ for SWNTs. We can obtain *ds* value is about 1.3 nm from the most-intense RBM peak ($\omega_{\text{RBM}} = 182.3 \text{ cm}^{-1}$), which is similar with the value given by the supplier. The inset in Fig. 1 is the micrograph of the SWNT/PVA composite, indicating high purity and uniform.



Fig. 1. (a) The Raman spectrum of the SWNT/PVA film. (b) The linear transmittance of the SWNT/PVA and pure PVA films. The pink rectangular denotes the operating wavelength region around 1060 nm.

Figure 1(b) shows the transmittance of pure PVA films and SWNT/PVA films measured by an UV-Visible-NIR spectrophotometer (Agilent Technologies, Cary 5000). The absorption at 1 μ m is mainly due to S2 transition of CNTs [31]. The modulation depth (MD) of SAs typically decreases with detuning from the resonant absorption wavelength [32]. It can be noted that the high MD of the nonlinear response is expected near 1 μ m (our operation wavelength), which corresponds to the first absorption peak in the transmittance measurement results in Fig. 1(b). We can also see that there is another larger absorption around 2 μ m (Fig. 1(b)), which indicates that the prepared SWNT SA can be potentially applied in a long wavelength regime.

Optical limiters show lower transmittance with high input laser density. OL effect can be attributed to many passive limiting mechanisms such as nonlinear absorption (multiphoton absorption, ISA), nonlinear light scattering, and nonlinear refraction (electronic or thermal effects) [33,34]. SWNT suspensions have been reported to have optical limiting effects [35]. Figure 2(a) shows the schematic setup of the power-dependent transmittance measurements. A home-made ps mode-locked Yb-doped fiber (YDF) laser is used as input laser source (center wavelength of 1060 nm, spectral width of around 8 nm, repetition rate of 23.2 MHz, and pulse duration of 7.6 ps). The incident optical power to the SWNT samples can be varied by changing the amplifying stage pump powers. An optical coupler is used to split the input beam into 30% reference signal and 70% input light to our samples. A two-channel power

meter (Ophire, PD300R-IR) is used to measure the output power simultaneously. The powerdependent transmittance of the SWNT/PVA films is shown in Fig. 2(b). The blue dot and the red curve represent the experimental result and the fitting result, respectively. A relative high MD of ~9.37% is measured at this wavelength. The saturation intensity is about 21 MW/cm², the nonsaturable absorption loss is about 28.72%. However, the SWNT/PVA films turned into the OL regimes at higher input power level (> 100 MW/cm²). The transmittance becomes lower with further increasing of the input powers (i.e., OL), which is mainly due to nonlinear scattering, two-photon absorption (TPA) effect, and other nonlinear absorption effects.



Fig. 2. (a) The schematic diagram of power-dependent transmittance measurements. (b) Nonlinear transmittance of the SWNT/PVA film.

Considering the inverse of the saturable absorption at high power density, we combine a SA coefficient and the TPA coefficient. Thus the total absorption coefficient can be written as a function of input intensity (I) [36,37]

$$\alpha(I) = \alpha_0 \frac{1}{1 + I/I_s} + \alpha_{NS} + \beta I \tag{1}$$

where the first term represent SA effect and the third term describes positive nonlinear absorption (NLA) such as TPA. α_0 is the linear absorption coefficient. α_{NS} is the nonsaturable absorption. *I* and *I_s* are laser radiation intensity and saturation intensity, respectively. β is TPA

coefficient. At high pump power, the transmittance of SWNT/SA become low. The transmittance decreases to 72%, as the pump power reaches 20 mW. Z-scan measurements can induce a problem, which was assumed to attribute the measured saturable-absorption characteristic to the hole of SA after being damaged [38]. Power-dependent measurement method, though is not as accurate as Z-scan method, can avoid this assumption.

Figure 3 shows the schematic diagram of the YDFL which incorporates SWNT absorber in the cavity. A 0.8-m-long Yb-doped fiber (YDF) with absorption coefficients of 500 dB/m at 976 nm is used as the gain medium. The Yb-doped fiber is pumped by a 976-nm laser diode through a wavelength division multiplexing (WDM) coupler. Other fibers in the cavity used in the cavity are HI-1060 fiber. In order to achieve mode-locking by SWNTs and make the light propagating unidirectionally, a polarization-independent isolator (PI-ISO) is inserted in the cavity. A polarization controller (PC) is mounted on the passive fiber to achieve certain polarization state in the cavity which may affect the absorption characteristic of SWNTs. A 90:10 output coupler is used to output the signal for measurement. To ensure stable modelocking in our all-normal-dispersion fiber cavity, a spectral filter needs to be introduced [39]. We integrate the fiber laser with 1064-nm filter with bandwidth of 10-nm, which induces the spectral filtering effect. Our SWNT film is cut into small pieces and sandwiched between two FC/PC fiber connectors. The fiber laser is pumped by a 976-nm laser diode (LD). The total length of our fiber cavity is about 9.56 m. The output spectra are detected by the optical spectral analyzer (OSA, AO-6315A). And the pulse trains are monitored by an oscilloscope with bandwidth of 6 GHz (LeCroy SDA) together with a high speed photodetector with bandwidth of 10 GHz (Kangguan). The mode-locked pulse width is measured by a commercial autocorrelator (APE pulseCheck 1600).



Fig. 3. Schematic diagram of the proposed fiber laser based on a SWNT SA.

3. Experimental results and discussion

Self-started mode-locking can be achieved by increasing the pump power to 47 mW. A typical spectrum in the log scale is shown in Fig. 4(a) and the spectrum in the linear scale is shown in Fig. 4(b) with a step-edge spectrum. The spectral width is about 0.17 nm which is smaller than the step edge-to-edge width of 0.6 nm. The corresponding oscilloscope trace is shown in Fig. 4(c), which indicates the proposed fiber laser operating at the repetition rate of 21.5 MHz corresponding to the cavity length. And the corresponding autocorrelation trace is shown in Fig. 4(d) with the pulse duration of about 317 ps, if a Gaussian profile is assumed. The output power is about 1.5 mW.



Fig. 4. Typical mode-locked state of the proposed fiber laser when the pump power is about 47 mW, (a) the spectrum in log scale and (b) the linear scale, (c) the corresponding oscilloscope trace with the single pulse (Inset), (d) corresponding autocorrelation trace with experimental and fitted results.

By adjusting the polarization controller, different types of spectra can be obtained. This is because adjustment of polarization controller alter not only the characteristics of saturable absorption of the SWNT, but also the phase delay of the cavity, which leads to different mode-locked states [40]. We find that there is no mode locking despite adjusting the PC in the fiber cavity, when the SWNT/PVA SA is removed from the fiber laser cavity. So the NPR effect is very weak in the cavity.

In addition, Q-switched states can be obtained, when the pump power increases above 80 mW. The repetition rate and the pulse duration can be varied by increasing the pump power. Figures 5 (a) and (b) show the typical optical spectrum and the corresponding oscilloscope trace, respectively, when the pump power is about 80 mW. The spectral width becomes narrower than the one in mode-locked states, and the interpulse-interval is 26.62 μ s that is far larger than the cavity round trip time of 46.5 ns, which indicates that the proposed fiber laser operate in the Q-switched states.



Fig. 5. (a) Typical spectrum of Q-switched operation when the pump power is about 80 mW. (b) Corresponding oscilloscope trace.

Figure 6(a) shows the pulse-train evolution by increasing pump power. Figure 6(b) shows the pulse duration and the repetition rate as a function of the pump power, respectively. Larger pump intensity could lead to smaller interpulse-interval (i.e., higher repetition rate) and smaller pulse width. As the pump power increases from 80 mW to 120 mW, the repetition rate varies from 30 kHz to 50 kHz, and the corresponding pulse width decreases from 2.7 μ s to 1 μ s. Further increasing the pump power would lead to optical power induced thermal damage of the SWNT composite. When the pump power is 120 mW the output power is about 988 μ W (corresponding to the pulse energy of 18.38 nJ), smaller than the output power of typical mode-locked output when the laser is pumped with lower power. This is mainly due to the OL effect of SWNT, which contributes higher intracavity loss.



Fig. 6. (a) The pulse train evolution versus pump power. The repetition rate becomes larger and the pulse duration decreases with the increase of the pump power. (b) The repetition rate and pulse duration versus the pump power, respectively. The green curve shows the pulse duration versus the pump powers, and the blue curve shows the repetition rates versus the pump powers.

Conventionally, a fiber laser operates in the Q-switched mode-locked states at the lower pump power, while it operates in the mode-locked states when the pump power is higher. However, due to the OL effects of the SAs, the proposed fiber laser can operate in the mode-locked states when the pump power is low (typically 47-80 mW), while it operates in the Q-switched states when the pump power is high (typically 80-120 mW). This is quite different from the former experiments observed. We believed that the SWNT SA performed like a mode locker when the pump power is low, and it performs as a Q switcher when the pump power is high enough that the OL effect dominates the absorption of SWNT/PVA film. So there exists a Q-switched effect formed in the laser cavity.

Q-switched laser has long pulse duration, which is suitable for laser marking and surface enhancement, while mode-locked laser has relatively narrow pulse, which is suitable for micro-machining application. By optimizing the inversed operating seed laser and implementing more amplifying stages, the inverted operation laser system can be used in the versatile material processing with alternate machining precision. The mode-locked and Qswitched fiber laser can also be potentially applied in optical communication and radar systems, respectively.

4. Conclusion

In conclusion, we demonstrate an Yb-doped fiber laser based on SWNT/PVA SA. The SA as well as the OL of SWNT/PVA is investigated. We firstly demonstrate that the mode locking can be obtained at low pump power and then Q-switching at high pump power. Dissipative solitons with peak spectral profile are generated in the cavity when the pump power is lower. Mode-locked state occurs at a lower pump power and Q-switched state occurs at a higher pump power. The SWNT/PVA films experiences OL effect at high power level, which plays an important role to obtain the inversed operation state in Yb-doped fiber lasers. The reversed operating fiber laser may be found many great potential applications.

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