Concentration effect of carbon nanotube based saturable absorber on stabilizing and shortening mode-locked pulse

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Abstract: We comprehensively investigated the concentration effect of dispersed single-walled carbon nanotubes (SWCNTs) in polymer films for being a saturable absorber (SA) to stabilize the mode locking performance of the erbium-doped fiber laser (EDFL) pulse through the diagnosis of its nonlinear properties of SA. The measured modulation depth was from 1 to 4.5% as the thickness increased 18 to 265 μm. The full-width half-maximum (FWHM) of the stable mode-locked EDFL (MLEDFL) pulse decreased from 3.43 to 2.02 ps as the concentrations of SWCNTs SA increased 0.125 to 0.5 wt%. At constant concentration of 0.125 wt%, the similar pulse shortening effect of the MLEDFL was also observed when the FWHM decreased from 3.43 to 1.85 ps as the thickness of SWCNTs SA increased 8 to 100 μm. With an erbium-doped fiber length of 80 cm, the shortest pulse width of 1.85 ps were achieved at 1.56μm with a repetition rate of 11.1MHz and 0.2 mW of the output power under an output coupling ratio of 5%. An in-depth study on the stable mode-locked pulse formation employing SWCNTs SA, it is possible to fabricate the SWCNT films for use in high performance MLEDFL and utilization of many other low-cost nanodevices.

OCIS codes: (140.4050) Mode-locked lasers; (140.3560) Lasers, ring.

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1. Introduction

Passively mode-locked erbium-doped fiber laser (MLEDFL) enables to generate optical pulses in picosecond and femtosecond regimes [1,2] and is often initiated by nonlinear optical elements with intensity-dependent response to favor optical pulsation over continuous–wave lasing [3]. The optical nonlinear element may be either a semiconductor saturable absorber mirror (SESAM) [1] or a nonlinear optical mirror [4]. The SESAMs have been widely used in ultrashort-pulse EDFLs for passive mode-locking [3,5]. However, the fabrication of SESAMs is suffered from a relatively cost-ineffective and time-consuming process in the complicated epitaxial systems, such as MBE and MOCVD, which usually requires a further ion-implanted treatment to shorten the carrier lifetime of the SESAM based saturable absorber. Recently, the single-wall carbon nanotubes (SWCNTs)-based saturable absorbers (SWCNTs SA) has been developed by a simple spin or spray coating process [6–10]. Because the absorption band of the SWCNTs can be shifted by varying their diameters to operate a broad spectral range, the SWCNTs enable the fabrication of a broadband saturable absorbing element with ultrashort recovery time [6]. Therefore, the SWCNTs SA may be a potential candidate for semiconductor-based ultrafast passive mode lockers and limiters.

Despite numerous studies on passively MLEDFLs using the SWCNTs SA as a nonlinear optical element [6–10], limited information is available for the processing parameter and the characterization of SWCNTs in fabricating films for stable MLEDFL pulse formation. In this study, we comprehensively investigated the concentration effect of dispersed SWCNTs in polymer films for being a SA to stabilize the mode locking performance of the EDFL pulse through the diagnosis of its nonlinear properties of SA. The SWCNTs SA is inserted in the EDFL ring to analyze correlation of stable MLEDFL pulse formation with different concentrations and thicknesses. The pulse trace and optical spectrum are characterized to the MLEDFL pulse formation. The condition of SWCNTs concentration and film thickness for optimizing mode-locking is also characterized and discussed. An in-depth study on the stable mode-locked pulse formation employing SWCNTs SA will broadly benefit to the utilization of the SWCNTs SA for use in many other low-cost photonic applications.

2. Methods

The SWCNTs SA has the ability of pulse shaping due to its nonlinear absorption properties. In general, the single-pass optical transmittance of SWCNTs SA, T(I), can be expressed as Eq. (1)

$$T(I) = \exp\left(-[\alpha(I) + \alpha_{ns}]L\right), \tag{1}$$

where L, α(I) and α_{ns} are the thickness, nonlinear absorption coefficient and non-saturation absorption coefficient of SWCNTs SA, respectively. The intensity dependent α (I) is given by Eq. (2)

$$\alpha(I) = \alpha_0 (1 + \frac{I}{I_{sat}})^{-1}, \tag{2}$$

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where the $\alpha_0 = \sigma N$ and $I_{sat} = \hbar\omega_l / \sigma\tau_{rex}$ are the linear absorption coefficient and saturation intensity, respectively. The σ, N, ω_l, and τ_{rex} are the absorption cross-section of SWCNT, concentration of SWCNTs SA, laser frequency, and energy relaxation time of SWCNT, respectively. According to the Eq. (1) and Eq. (2), the performance of the MLEDFL may correlate to the concentration and thickness of SWCNTs SA. The purpose of this work is to investigate the parameter dependence on mode-locking pulse formation, such as the SWCNT concentration in films and the film thickness.

The SWCNT material was purchased from Golden Innovation Business Company. Based on the corresponding van Hove singularity [11,12], the diameter distribution of the SWCNTs was chosen from 1 to 1.5 nm to match the EDFL ring operating at 1550 nm. The purity was greater than 90%.

The water-soluble polyvinyl alcohol (PVA) of high molecular weight was used as the substrate for films. Since the SWCNT was not dispersible in water, the surfactant of sodium dodecylbenzenesulfonate (SDBS) was used to assist the SWCNT dispersion [13,14]. The process of fabricating the SWCNTs SA involved four steps. The SWCNTs and SDBS dispersant were mixed in the water to form the finely dispersed solution by using ultrasonic vibrator for 3 hours to uniformly disperse the SWCNTs at an appropriate weight ratio. The amounts of SWCNTs varied for making up different concentrations in SWCNTs SA. The PVA in water solution was added and stirred for 6 hours. The viscous SWCNTs-PVA solution was spin-coated or just dropped on the glass substrate to form the SWCNTs-PVA film with a uniform thickness. The thickness of the film can be adjusted by changing the speed of the spin coater. After evaporating and drying at ambient temperature, the SWCNTs SA films were formed.

The linear optical absorption of the SWCNTs SA was examined by using UV-Visible-NIR spectrophotometer. From the location of absorption peak, the SWCNTs SA was within the operating wavelength of EDFL ring near 1550nm. The alpha-step profilometer was used to measure the thickness of SWCNTs SA. The nonlinear transmission was evaluated through the measurement of single-pass optical loss by using the ML laser at 1560 nm central wavelength. The intensity level from 1 to 100 MW/cm^2 was used for the nonlinear transmission measurement.

Figure 1 shows an experimental setup of the passive MLEDFL ring. A 980 nm diode laser was used to pump 80-cm length of erbium-doped fiber (EDF) as the gain medium. The peak absorption of EDF was 80 dB at 1530 nm and the maximum pumping power was 120mW at 980 nm. The SWCNTs SA placed between two FC/APC fiber connectors was integrated into the EDFL ring to generate the MLEDFL pulses. An isolator was employed to prevent backreflection and to ensure the unidirectional operation. The emission light passed the SWCNTs SA and launched into EDFL ring through a 95% output coupler. A 3 dB power splitter was connected to a 5% output port for simultaneous observing the pulse train and optical spectrum through the oscilloscope and optical spectrum analyzer, respectively. The EDFL output transits from cw to pulse mode by increasing the pumping power level. A stable pulse train is achieved with the aid and detune of an intra-cavity PC. In this study, we focus on evaluating the temporal and spectral characteristics of EDFL pulses from varying concentration or thickness of SWCNTs SA without optimizing chirp-free shortest pulse generation and keeping the other parameters of the EDFL ring unchanged during experiment. Figure 2 shows the pulse train of MLEDFL ring. The repetition rate of MLEDFL ring was 11.11MHz. The pulse trace of output was captured from autocorrelator after being extra-amplified by EDF amplifier.

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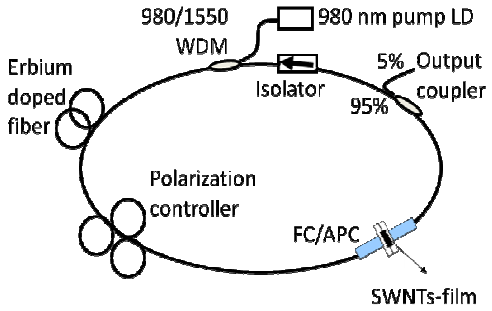

Fig. 1. Experimental setup of EDFL ring incorporating SWCNTs SA.

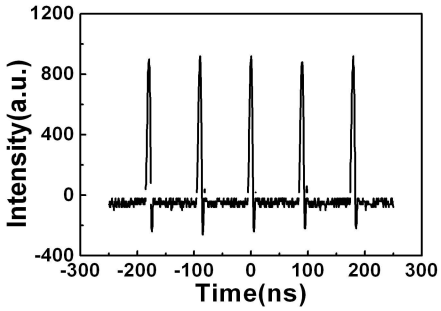

Fig. 2. The pulse train of mode-locked EDFL ring output observed from oscilloscope

3. Results and discussions

In order to examine the dispersion of SWCNTs in polymer films, a metallographic analysis was used to observe the microstructures of the SWCNTs in film and to compare the SWCNTs SA film with different concentrations. A 50 time magnification of optical microscope exhibited the microstructure of the SWCNTs SA under the different concentrations of 0.5 and 1 wt%, as shown in Fig. 3. In Fig. 3(a), there was none of obvious aggregation of SWCNTs at the concentration of 0.5 wt%. The re-aggregation phenomenon appeared in the SWCNTs SA when the SWCNT concentration was up to 1 wt%, as shown in Fig. 3(b).

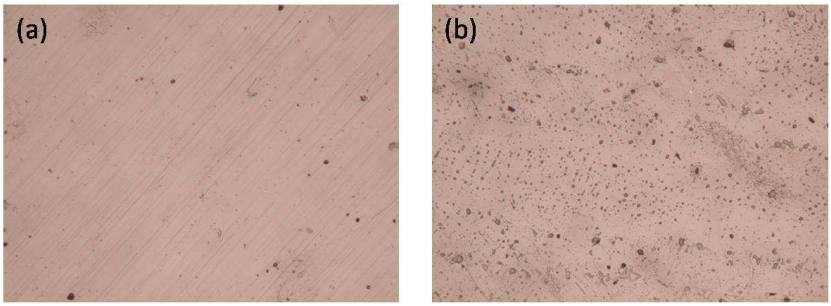

Fig. 3. Metallographic photos of SWCNTs SA under concentrations of (a) 0.5 and (b) 1 wt%.

To probe the operating wavelength of SWCNTs SA, a UV-Visible-NIR spectrophotometer was used to measure the linear optical absorption of SWCNTs-PVA film within 400-1800 nm wavelength, as shown in Fig. 4. It showed that the peak absorption wavelength of SWCNT-PVA film corresponding to first van Hove was near 1550 nm, locating at the erbium gain window.

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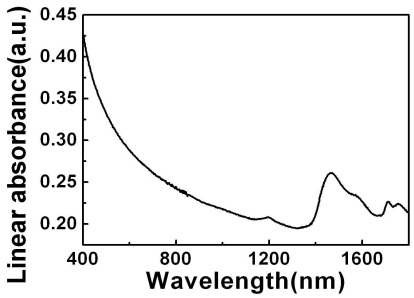

Fig. 4. Linear optical absorbance of SWCNTs SA.

Figure 5 shows the nonlinear transmission measurement with 100 μm thickness of the SWCNTs SA. According to Eq. (1) and Eq. (2), the modulation depth, saturation intensity and nonsaturable losses were estimated about 4%, 18 MW/cm^2 and 44.5%, respectively, based on the experimental data fitting. The saturation intensity for SAs with different thicknesses ranging from 18 to 265 μm were measured within 18 ~19 MW/cm^2 indicating the high purity and well dispersed SWCNTs sample are prepared.

The modulation depth and nonsaturable losses are sensitive to the thickness of SWCNTs SA, as shown in Fig. 6. The modulation depth increased as the sample thickness increased. It implies that better pulse shaping is occurred with thicker saturable absorber. Except for the increasing connection loss with larger gap between fiber end surfaces, the transmission profile shifts down with increasing thickness. It inevitably causes a larger threshold pumping power to achieve a mode-locking operation.

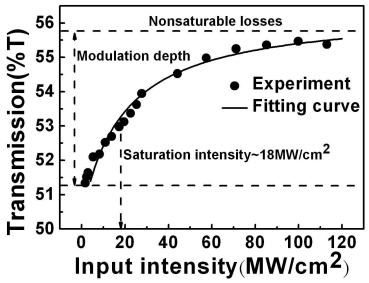

Fig. 5. Nonlinear transmission as a function of input intensity with 100 μm thickness of the SWCNTs SA.

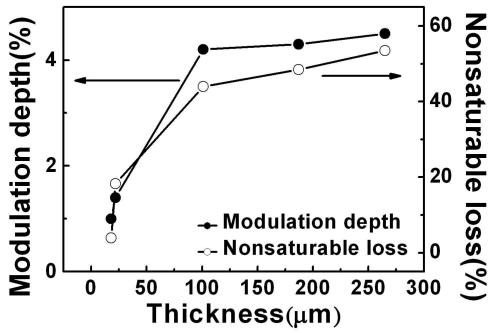

Fig. 6. Modulation depth and nonsaturable losses versus thickness of the SWCNTs SA.

After the setup of fiber ring laser, the gain and the whole cavity length remain unchanged. When the different saturable absorbers were used, the mode-locking performances including pulse trace and optical spectrum are monitored. During measurement, a stable Q-switched

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pulse-train was observed by increasing the pump power beyond the threshold Q-switching condition. As the pump power increases, the spectrum becomes broader and the pulse width becomes shorter concurrently. Further increasing the pumping power to the mode-locking threshold, the multiple-pulse generated within the Q-switching envelope can be observed. The mode-locking condition can easily be achieved when increasing the thickness of the SWCNTs SA to >100 μm. The change between Q-switching and mode-locking conditions can also be controlled via the detuning of the intracavity polarization controller. The dependence of pumping power required to reach mode-locking threshold plotted as a function of the saturable absorber thickness is shown in Fig. 7. The transition zone from CW to ML threshold region became broader as the thickness increases.

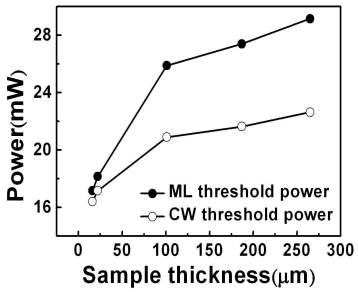

Fig. 7. CW and ML threshold pump power versus thickness of the SWCNTs SA.

In order to investigate the effect of concentration and thickness of SWCNTs SA on mode-locked pulse formation, concentrations from 0.15 to 0.5 wt% and thicknesses from 8 to 265 μm of SWCNTs SA were used. The inset of Fig. 8 shows the optical spectrum of EDFL output under 103 mW pumping power with 0.15 wt% concentration and 8 μm thickness of SWCNTs SA. The FWHM of output pulse trace were measured 3.43 ps from the autocorrelator. The average output power is 0.88mW. This pulse trace is not stable and is still not the well fitting hyperbolic secant squared pulse profile, as shown in Fig. 8. This situation indicates the on-set of the MDEDFL operation.

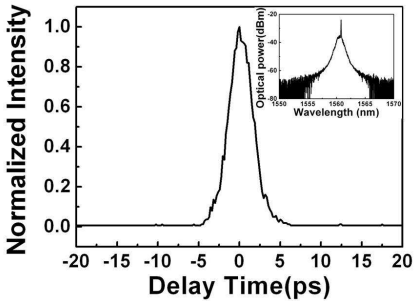

Fig. 8. Autocorrelator trace and optical spectrum (inset) of MLEDFL with 0.125 wt% concentration and 8 μm thickness of the SWCNTs SA.

The inset of Fig. 9 shows a broader optical spectrum under the same pumping power of 103 mW when the concentration of SWCNTs SA increases to 0.5 wt% and the thickness is kept at 8 μm. The optical spectrum was centered at 1560.5 nm with a 3-dB spectral bandwidth of 2.2 nm. The average output power is 0.8 mW. A fitting hyperbolic secant squared pulse profile is shown in Fig. 9. The FWHM of pulse and time-bandwidth product were 2.02 ps and 0.548, respectively. In comparison with 0.125 wt% concentration of SWCNTs SA in Fig. 8, Fig. 9 shows that the pulse trace is more stable in MLEDFL operation.

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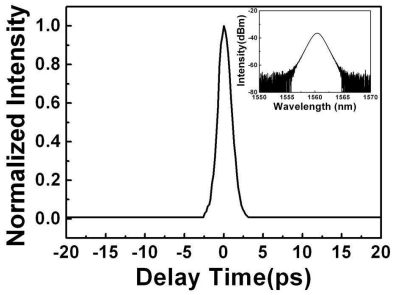

Fig. 9. Autocorrelator trace and optical spectrum (inset) of MLEDFL with 0.5 wt% concentration and 8 μm thickness of the SWCNTs SA.

By maintaining the same concentration of 0.125 wt% and increasing the film thickness to 100 μm, the optical spectrum and pulse trace were measured under the same pumping power of 103 mW. The inset of Fig. 10 shows the symmetric side spikes in optical spectrum which is centered at 1558.7 nm with a 3-dB spectral bandwidth of 4.15 nm. It implies a soliton-like pulse operation [15], as shown in Fig. 10. For thicker SA and the larger gap between two connectors, the average output power decreased to 0.2 mW. The FWHM of pulse was measured 1.85 ps and was well fitting hyperbolic secant squared pulse profile. Comparing with 8 μm of SWCNTs SA in Fig. 8, the spectrum in Fig. 10 was relatively broad.

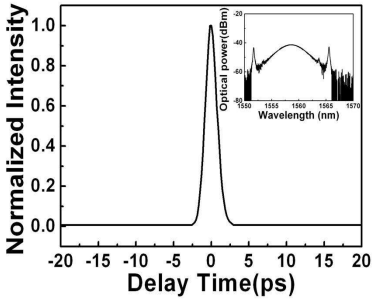

Fig. 10. Autocorrelator trace and optical spectrum (inset) of MLEDFL with 0.125 wt% concentration and 100 μm thickness of the SWCNTs SA.

In order to clarify the effect of concentration and thickness for the SWCNTs SA on the MLEDFL pulse, the FWHM of pulse width and 3-dB spectral bandwidth versus the concentrations and thicknesses are investigated. The FWHM of pulse width decreased from 3.43 to 2.02 ps as the concentration of SWCNTs SA increased 0.125 to 0.5 wt% while the thickness of SWCNTs SA is 8 μm, as shown in Fig. 11(a). For a short pulse width of 2.02 ps, a broader 3-dB spectral bandwidth of 2.2 nm was obtained at the concentration of 0.5 wt%. This indicates that the MLEDFL pulse becomes short as the concentration of SWCNTs SA increases. However, the pulse width of the passively MLEDFLs broadened to 2.45 ps as concentration increased to 1 wt%. The reasons are attributed to the reaggregation of SWCNTs with increasing concentration and the extremely denseness which greatly attenuates the circulation laser power inside the EDFL ring to degrade or restrict the initiation of mode-locking in the EDFL. Figure 11(b) shows that the FWHM of pulse width decreases from 3.43 to 1.85 ps (or 3-dB spectral bandwidth increases from 1.25 to 4.15 nm) as the thickness of SWCNTs SA increases 8 to 100 μm while the concentration of SWCNTs SA is 0.125 wt%. The pulse width became stable as thickness of SWCNTs SA increased 100 to 265 μm. In addition, the time-bandwidth product obviously changed from about 0.5 to 0.9 when the thickness increased to 100 μm. It implies that the pulse is chirped in the ring cavity when we change the saturable absorber with different thicknesses. The chirping of pulse may contribute to the resonant SWCNTs SA dispersion enhancement as the thickness of SWCNTs

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SA increases. From Fig. 11, the effects of the SWCNT concentration and the SWCNTs SA thickness on the stabilization and shortening the MLEDFL pulse width are clearly realized.

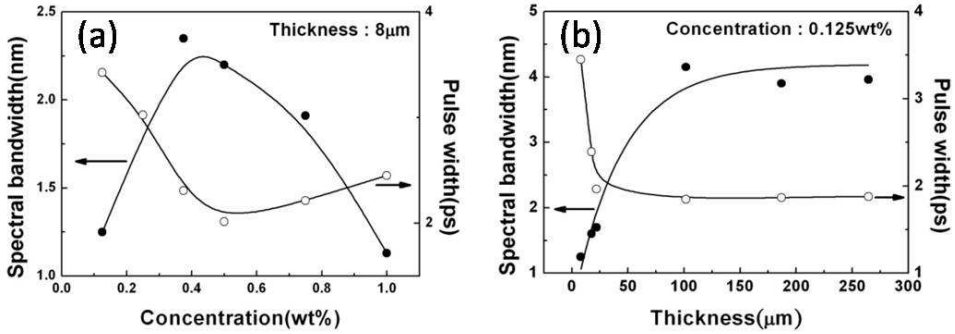

Fig. 11. FWHM of pulse width and 3-dB spectral bandwidth of MLEDFL versus (a) concentration (b) thickness of SWCNTs SA.

The nonlinear absorption diagnosis of different saturable absorbers was used to analyze the concentration effect in more detail. When the modulation depth is small, the pulse trace accompanied with longer pulse width is unstable and the spectral bandwidth broadening is limited. When the modulation becomes large, the well stable fitting hyperbolic secant squared pulse profile accompanied with shorter pulse width can be obtained and the spectral bandwidth can be expanded to 4.15 nm. Besides, the increasing nonsaturable losses and high chirping phenomenon were observed for the cause of using SA with larger thickness.

Figure 12 shows TBP as a function of ML ring laser cavity length by changing single-mode fiber length. The result revealed the dependence between pulse qualities and cavity length and the best pulse quality obtained with single-mode fiber length of around 16m. It can be attributed to dispersion compensation from single-mode fiber since mode-locking threshold and output power are almost fixed. This further suggests a guideline of cavity length as designing a mode-locked fiber laser with SWCNT as saturable absorber.

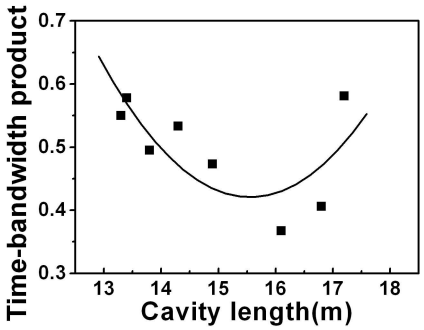

Fig. 12. Time-bandwidth product as a function of ML ring laser cavity length

4. Conclusion

In conclusion, the concentration of SWCNTs in the polymer film with changing thickness was characterized to understand their effects on stability and pulse width of the MLEDFL through the diagnosis of its nonlinear properties of SA. At constant 8-μm thickness of SWCNTs SA, the FWHM of the pulse width for MLEDFL decreased from 3.43 to 2.02 ps as the concentration of SWCNTs SA increased 0.125 to 0.5 wt%. For a short pulse width of 2.02 ps, a broader 3-dB spectral bandwidth of 2.2 nm was obtained at the concentration of 0.5 wt%. Apparently, the EDFL pulse was stabilized and shortened as the concentration of SWCNTs SA increased to 0.5 wt%. At constant 0.125 wt% concentration of SWCNTs SA, the measured modulation depth was from 1 to 4.5% as thickness increased 18 to 265 μm. The

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EDFL pulse width was shortened from 3.43 to 1.85 ps or the EDFL 3-dB spectral band was increased from 1.25 to 4.15 nm as the thickness of SWCNTs SA increased 8 to 100 μm. However, the associated nonsaturable losses from 4.0 to 53.5% also inevitably increased the ML threshold power and suppressed the broadening of the ML spectral bandwidth to limit the pulse width within a finite range. In addition, the chirping of pulse also changed with different thicknesses of the SWCNTs SA. For the stability of ML generation under same laser configuration and beam size, the concentration of 0.5 wt% in the range of film thickness of 22-100 μm exhibited a stable MLEDFL pulse formation. Detailed understanding on the stable mode-locked pulse formation employing SWCNTs SA, it is possible to fabricate the SWCNT films for use in high performance and low-cost MLEDFLs and utilization of many other low-cost nanodevices.

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