

# Mode locking of a fibre laser with a matrix-less carbon nanotube film

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## ABSTRACT

This work for the first time reports experimental results on Er fibre laser mode-locked with saturable absorbers based on single-walled matrix-less carbon nanotube films fabricated by aerosol method. Nearly transform-limited output pulses had duration of 1.7 ps and average power of 0.5 mW. Measured high-contrast RF output spectra (noise suppression over 70 dB in 1-GHz band) confirm high quality of achieved mode locking. Discussed are details of the aerosol method of matrix-less carbon nanotube film fabrication and parameters of studied mode-locked fibre laser. Application of matrix-less carbon nanotube films for mode locking and Q-switching of fibre and other laser types are analysed.

**Keywords:** carbon nanotube, mode-locked fibre laser, saturable absorber, matrix-less carbon nanotube film

## 1. INTRODUCTION

Materials made of low-dimensional structures may be efficiently used as saturable absorbers in lasers. For instance, carbon nanotubes (CNT) have already demonstrated their potential as saturable absorbers for ultra-fast fibre lasers [1–6]. Conventionally, CNT are used in the form of a polymer film with single-wall carbon nano-tubes uniformly distributed over its volume [1, 7]. Interaction of laser radiation with such a film doped with CNT is often provided by clamping it between two optical fibre ferrules of a fibre-optical connector or by applying a coating of such film onto either a transmission or reflection surface of an optical element within the fibre laser cavity. However, this method suffers from the optical properties of the host polymer changing under the combined action of ultra-short laser pulses and the ambient conditions. This results in unstable laser radiation parameters and eventual degradation of the polymer [8, 9]. In order to overcome this issue, various attempts were made to deposit pure CNT (without a polymer matrix) onto optical surfaces crossed by the beam [10–13] or interacting with laser radiation [14]. However, in their large majority, these methods rather represented exotic laboratory exploration of the future potential far from technologically complete and practical approaches.

In recent years, a new technology of aerosol chemical vapour deposition (CVD) based fabrication of free-standing single-walled CNT films has been developed and successfully implemented [15]. Research into application of such films in lasers generates a good deal of interest since the technology used in their production is relatively well established, thus facilitating their practical utilisation and at the same time solving the challenge of gageing suffered by conventional CNT-based saturable absorbers.

The present work reports the results of studies of an Er fibre laser mode-locked with saturable absorber based on single-walled polymer-free CNT film fabricated by aerosol method.

## 2. EXPERIMENT

The experimental installation layout is schematically shown in Fig. 1. The ring fibre laser only included polarisation-maintaining elements oriented identically for minimisation of the non-linear polarisation evolution effect [16, 17]. The 1-m long active Er-doped fibre was core-pumped in the direction opposite to the direction of laser radiation circulation set by a fibre Faraday rotator. A 30% fibre splitter was used to couple the intra-cavity radiation out of the cavity. A polymer-free single-walled carbon nanotube film with a 100- $\mu\text{m}$  thickness and linear transmission of 60% at 1.5  $\mu\text{m}$  was sandwiched between two ferrules of an FC/APC fibre connector.

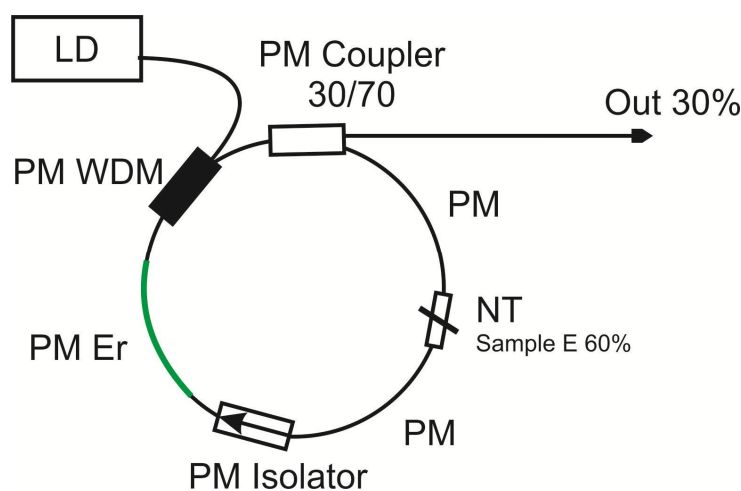


Fig. 1. Experimental set-up layout: LD – pump laser diode, WDM – wavelength division multiplexer, OC – output coupler, SA – saturable absorber sandwiched between two optical fibre ferrules: polymer-free single-walled carbon nanotube film, OI – optical isolator, EDF – Er-doped fibre.

CNT were synthesised by the aerosol (floating catalyst) CVD technique [21, 22]. The CNTs grow on the surface of iron particles suspended in gas phase inside a heated flow reactor and collected on a micro-porous filter. Deposited CNT films can be transferred from the filter to any other substrate by simple dry press transfer technique just by pressing it against the surface of the secondary substrate [23]. This process does not involve any liquid dispersion and purifications steps; also no polymer support is introduced in the film. Transmission electron microscopy confirmed formation of single-walled carbon nanotubes with an average diameter of 2.2 nm (Fig. 2(a)). The morphology of the CNT films used for fabrication of the saturable absorber is shown in Fig. 2(b). The linear transmission spectrum of the used CNT film is shown on the Fig. 2(c). The laser generation wavelength marked with an arrow corresponds to the wing of the S22 transition [24]. Since the studied samples only contained single-walled carbon nanotubes, these latter entirely defined the nonlinear optical properties of these samples.

The total cavity length was around 4 m and the cavity fibres had anomalous dispersion at the generation wavelength. The net dispersion of the cavity was  $-0.09 \text{ ps}^2$  (at 1,560 nm). The studied fibre laser had a 40-mW generation threshold, above which it entered mode-locked operation. The best generation stability was achieved at the pump power of 50 mW with the average output power of 0.5 mW. Higher pump radiation power led to respectively higher output power of up to 0.6–0.7 mW, but at the same time also to higher output power instability. Further increase of the pump power resulted in emergence of multi-pulse mode locking, in which a sequence of several pulses are propagating along the cavity. This mode-locked regime was not harmonic because the pulses were not uniformly spaced in time. The observed output beam quality was typical of fibre lasers based on single-mode fibres, and the corresponding beam quality factor  $M^2$  was close to unity.

Figure 3(a) shows the auto-correlation function of the generated pulses and their spectrum. The auto-correlation function was 2.6-ps wide, thus corresponding to pulse duration of 1.7 ps in the approximation of sech-shaped pulses. The auto-correlator used in these measurements (FS-PS-Auto, Tekhnoscan) had temporal resolution of 20 fs. The width of the optical spectrum presented in Fig. 3(b) is around 1.5 nm, which indicates that the generated pulses were close to transform-limited. The output radiation spectrum was registered with an optical spectrum analyser ANDO AQ6315, whose spectral resolution is as narrow as 0.05 nm. The pulse repetition rate was 50.3 MHz at the average output power of 0.5 mW. The pulse peak power was measured at 5.1 W and the pulse energy at 10 pJ.

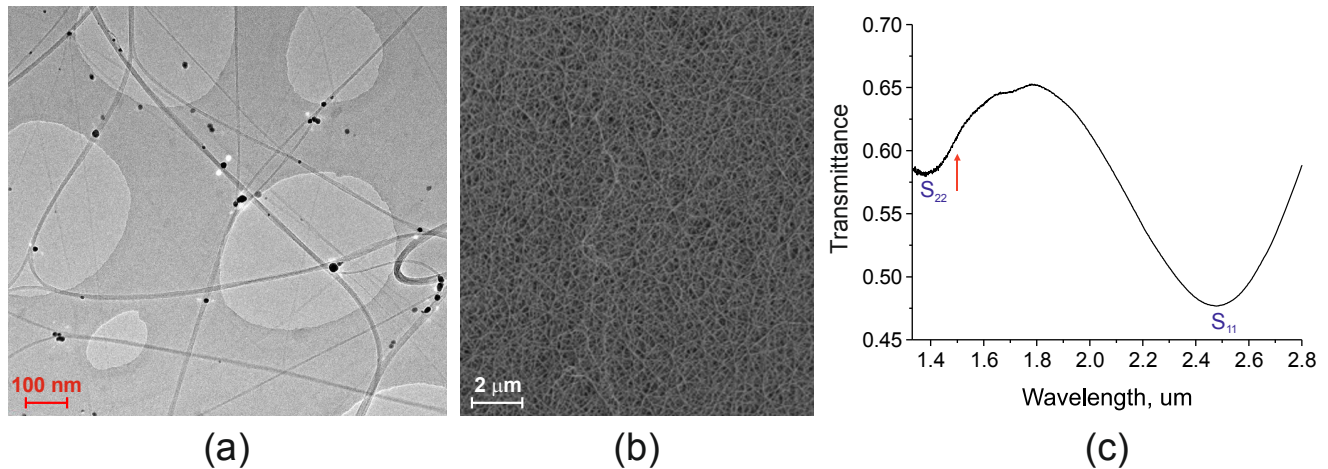


Fig. 2. (a) Transmission electron microscopy image of the CNT and (b) scanning electron microscope image of CNT film, (c) wavelength-dependent linear transmittance of the studied polymer-free CNT film:  $S_{11}$  and  $S_{22}$  – absorption bands corresponding to electronic transitions between van Hove singularities in the valence and conduction bands; the laser wavelength is marked with an arrow.

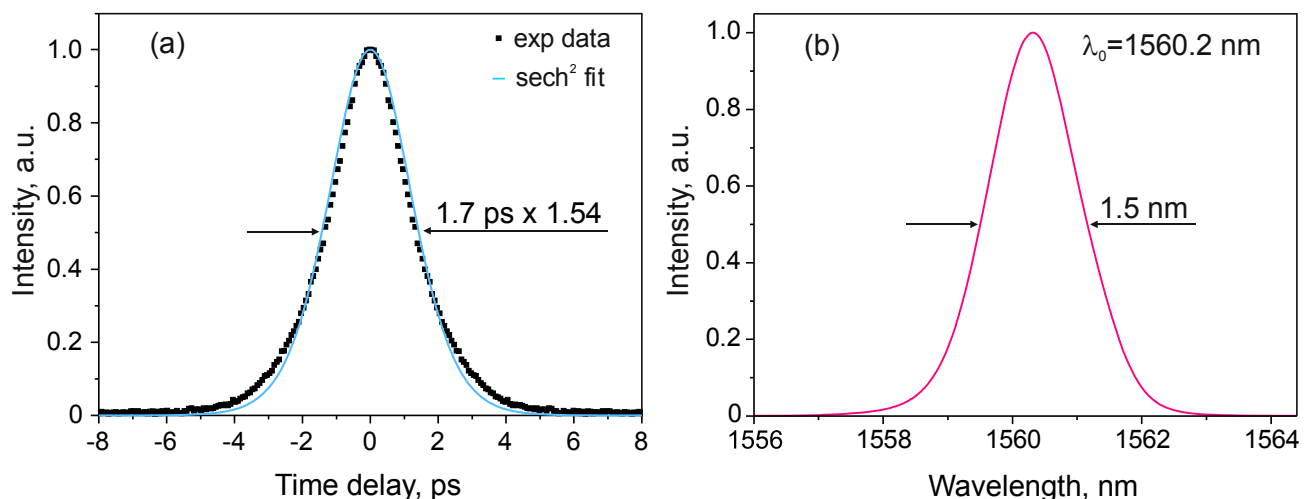


Fig. 3. Pulses generated with a polymer-free single-walled CNT film having linear transmission of 60%: (a) laser output auto-correlation function; (b) laser output spectrum at the average output power of 0.5 mW.

When examining the laser with a thinner sample of single-walled CNT film having 80% transmission at 1.5  $\mu\text{m}$ , the laser cavity was slightly longer (4.6 m), corresponding to the pulse repetition rate of 45.6 MHz. Overall, the generation parameters of the laser with this second sample were analogous to those with the first sample, but there were certain quantitative differences: the autocorrelation function was 6.8-ps wide (Fig. 4(a)), which corresponds to 4.4-ps duration of sech-shaped pulses. The optical spectrum width was equal to 0.8 nm (Fig. 4(b)), also indicating that the generated pulses were close to transform-limited. With this second CNT film sample, the optimal average output power amounted to 0.4 mW, corresponding to pulses with peak power of 1.7 W and energy of 8.8 pJ. As the pump radiation power was raised, the behaviour of the laser with this second CNT film was different. The laser entered a Q-switched regime accompanied with irreversible modification of the optical properties of the CNT sample because reduction of the pumping power did not lead to restoration of mode-locked operation either immediately or after power-cycling the pump laser. Q-switched generation was triggered at the peak radiation power density of 23  $\text{GW}/\text{m}^2$ .

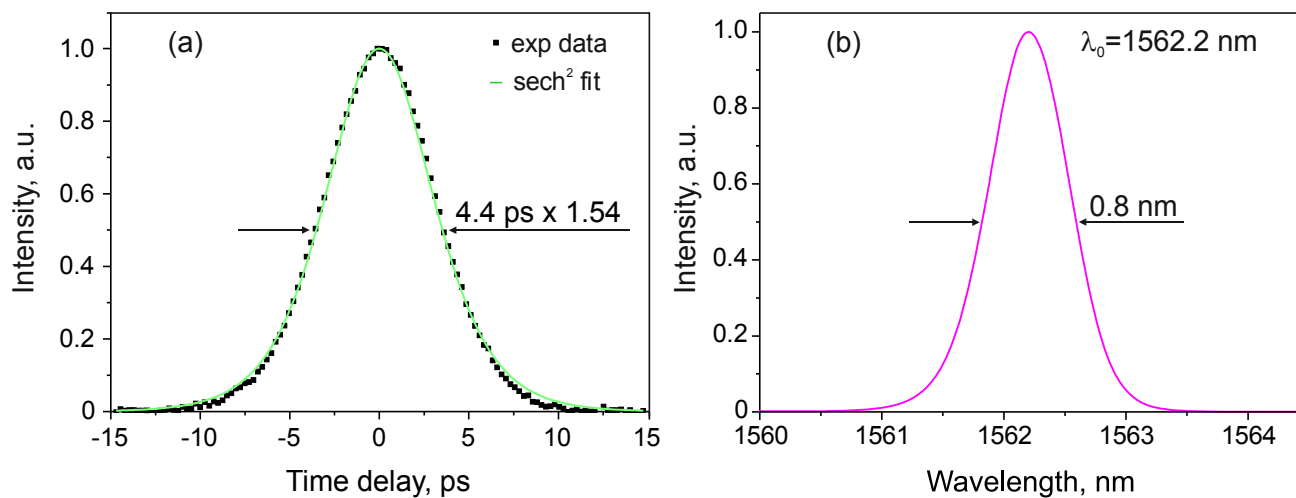


Fig. 4. Pulses generated with a polymer-free single-walled CNT film having 80% linear transmission: (a) auto-correlation function of the output pulses; (b) laser output spectrum at the average output power of 0.4 mW.

In order to evaluate the quality of the observed mode locking, we analysed RF spectra of the laser pulse train near the fundamental repetition frequency and its 20<sup>th</sup> harmonic (for the first CNT sample with 60% transmission) or 22<sup>th</sup> (for the sample with 80% transmission). All the registered RF spectra (Fig. 5) feature relatively high contrast exceeding 60 dB and no side peaks at both the fundamental frequency and its comparatively high harmonics. These spectra suggest high quality of mode locking provided by polymer-free single-walled CNT films.

One salient feature of the studied free-standing single-walled carbon nanotube films is their fairly high adhesion to various surfaces. They can be easily transferred from the filter onto practically any other substrate by a room-temperature dry-press transfer technique [15, 24]. The transfer process is extremely simple, and no dispersion or cleaning steps are needed prior to press transfer. To attach the CNT film onto a connector ferrule one needs simply to press with ferrule on the filter with a deposited CNT layer. This makes the process significantly faster, cheaper, and more environmentally friendly than the traditional liquid-based CNT deposition processes.

It is equally pertinent to note that essential properties of CNT themselves may also be affected by oxidation in air [18, 19] or elevated temperatures [20]. In order to avoid such modification, free-standing CNT films must be isolated from air (for instance, by placing them inside a sealed fibre connector, between whose ferrules a free-standing CNT film is clamped) and used in conditions ensuring that the laser radiation absorbed within the CNT film does not result in excessive heating of the film.

It is necessary to note that application of free-standing CNT films in fibre lasers whose cavities are totally polarisation-maintaining additionally offers a solution of the problem of triggering specific mode-locked regimes in these lasers [25, 26]. This is important, for instance, for implementation of fibre-optical super-continuum generators with specified parameters [27, 28] or fibre-based Raman sources of radiation [29, 30].

### 3. CONCLUSION

The presented work reports on study of all-fibre lasers mode-locked with a polymer-free carbon nanotube film. High-quality mode locking was achieved in a ring Er fibre laser with output pulse duration of 1.7 and 4.4 ps for the CNT film samples having respective transmission of 60% and 80% at 1.5  $\mu$ m. The generated pulses were close to transform-limited at the optimal output radiation power equal to 0.5 mW (the sample with linear transmission of 60%) and 0.4 mW (the sample with linear transmission 80%). Experiments conducted during this work demonstrate that free-standing single-walled carbon nanotube films may be used in lasers as saturable radiation absorbers not relying on traditional polymer matrices. Polymer-free carbon nanotube films are the foundation of the next generation of CNT-based saturable absorbers, whose optical properties do not suffer from degradation over time.

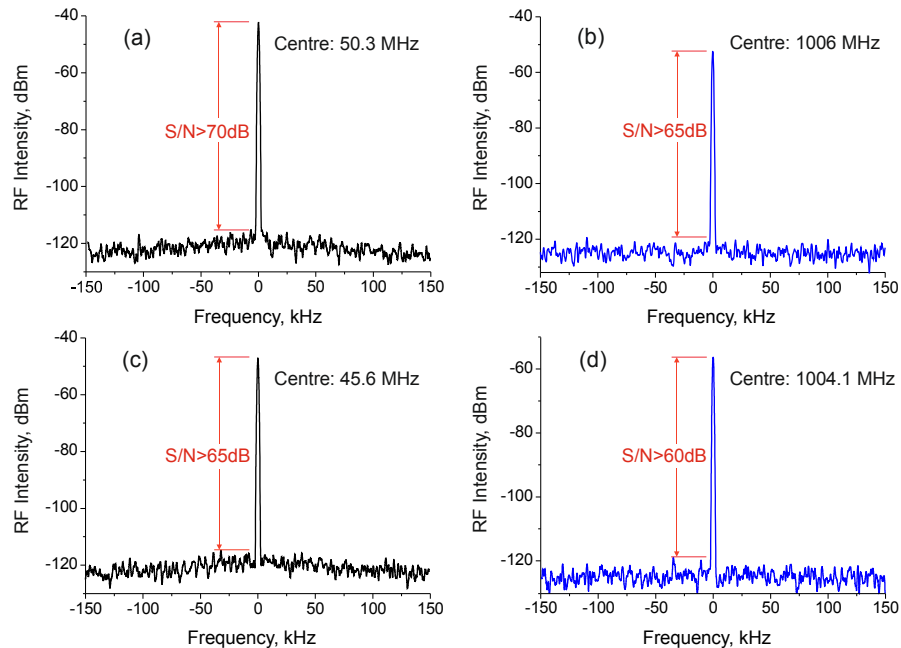


Fig. 5. RF spectra of the laser output for samples with linear transmission of 60% (a), (b) and 80% (c), (d): (a), (c) – RF spectra in the vicinity of the fundamental pulse repetition frequency; (b), (d) – RF spectra in the vicinity of the 20<sup>th</sup> (b) and 22<sup>th</sup> (d) harmonics of the fundamental pulse repetition rate.

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