# In-fiber microchannel device filled with a carbon nanotube dispersion for passive mode-lock lasing

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Abstract: Fueled by their high third-order nonlinearity and nonlinear saturable absorption, carbon nanotubes (CNT) are expected to become an integral part of next-generation photonic devices such as all-optical switches and passive mode-locked lasers. However, in order to fulfill this expectation it is necessary to identify a suitable platform that allows the efficient use of the optical properties of CNT. In this paper, we propose and implement a novel device consisting of an optofluidic device filled with a dispersion of CNT. By fabricating a microchannel through the core of a conventional fiber and filling it with a homogeneous solution of CNTs on Dimethylformamide (DMF), a compact, all-fiber saturable absorber is realized. The fabrication of the micro-fluidic channel is a two-step process that involves femtosecond laser micro-fabrication and chemical etching of the laser-modified regions. All-fiber high-energy, passive mode-locked lasing is demonstrated with an output power of 13.5 dBm. The key characteristics of the device are compactness and robustness against optical, mechanical and thermal damage.

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#### **References and links**

- 1. H. Kataura, Y. Kumazawa, Y. Maniwa, I. Umezu, S. Suzuki, Y. Ohtsuka, and Y. Achiba "Optical properties of single-wall carbon nanotubes," Synth. Met. 103, 2555-2558 (1999)
- Y.-C. Chen, N. R. Raravikar, L. S. Schadler, P. M. Ajayan, Y.-P. Zhao, T.-M. Lu, G.-C. Wang and X.-C. Zhang, "Ultrafast optical switching properties of single-wall carbon nanotube polymer composites at 1.55 µm," Appl. Phys. Lett. 81, 975-977 (2002)
- S. Y. Set, H. Yaguchi, Y. Tanaka and M. Jablonski, "Ultrafast Fiber Pulsed Lasers Incorporating Carbon 3. Nanotubes," IEEE J. Sel. Top. Quantum Electron. 10, 137 (2004).
- S. Yamashita, Y. Inoue, S. Maruyama, Y. Murakami, H. Yaguchi, M. Jablonski and S. Y. Set, "Saturable absorbers incorporating carbon nanotubes directly synthesized onto substrates and fibers and their application to mode-locked fiber lasers," Opt. Lett. 29, 1581-1583 (2004).
- 5. A. Martinez, S. Uchida, Y-W Song, T. Ishigure, and S. Yamashita, "Fabrication of Carbon Nanotube-Polymethyl-methacrylate Composites for Nonlinear Photonic Devices," Opt. Express 16, 11337-11343 (2008).
- Y-W Song, S. Yamashita, C. S. Goh, and S. Y. Set, "Carbon nanotube mode lockers with enhanced nonlinearity via evanescent field interaction in D-shaped fibers," Opt. Lett. 32, 148-150 (2007)
- 7. Y-W. Song, K. Morimune, S.Y. Set, and S. Yamashita, "Polarization insensitive all-fiber mode-lockers
- functioned by carbon nanotubes deposited onto tapered fibers," Appl. Phys. Lett. **90**, 021101 (2007) T. Oomuro, R. Kaji, T. Itatani, H. Ishii, E. Itoga, H. Kataura, M. Yamashita, M. Mori, and Y. Sakakibara, 8. "Carbon Nanotube-Polyimide Saturable Absorbing Waveguide Made by Simple Photolithography," in Conference on Lasers and Electro-Optics/Quantum Electronics and Laser Science Conference and Photonic Applications Systems Technologies, OSA Technical Digest Series (CD) (Optical Society of America, 2007), paper CThV1. Y-W Song, S. Yamashita, and S. Maruyama, "Single-walled carbon nanotubes for high-energy optical pulse
- 9. formation," Appl. Phys. Lett. 92, 021115 (2008)
- 10. D. Psaltis, S. R. Quake, and C. Yang, "Developing optofluidic technology through the fusion of microfluidics and optics," Nature 442, 381-386, (2006).
- 11. C. Monat, P. Domachuk, and B. J. Eggleton, "Integrated optofluidics: A new river of light," Nat. Photonics 1, 106-114 (2007).

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- Y. Lai, K. Zhou, L. Zhang, and I. Bennion, "Microchannels in conventional single-mode fibers," Opt. Lett. 31, 2559-2561 (2006)
- Y. Kondo, J. Qiu, T. Mitsuyu, K. Hirao, and T. Yoko, "Three-dimensional microdrilling of glass by multiphoton process and chemical etching," Jpn J. Appl. Phys. 38, L1146–L1148 (1999).
- A. Marcinkevicius, S. Juodkazis, M. Watanabe, M. Miwa, S. Matsuo, H. Misawa, J. Nishii, "Femtosecond laser-assisted three-dimensional microfabrication in silica," Opt. Lett. 26, 277–279 (2001).
- RR Gattass and E. Mazur, "Femtosecond Laser micromachining in transparent materials," Nat. Photonics, 2, 219-225 (2008)
- K. Zhou, Y. Lai, X. Chen, K. Sugden, L. Zhang, and I. Bennion, "A refractometer based on a micro-slot in a fiber Bragg grating formed by chemically assisted femtosecond laser processing," Opt. Express 15, 15848-15853 (2007)
- R. Osellame, V. Maselli, R. Martinez Vazquez, R. Ramponi and G. Cerullo, "Integration of optical waveguides and microfluidic channels both fabricated by femtosecond laser irradiation," Appl. Phys. Lett., 90, 231118 (2007).

## 1. Introduction

The nonlinear optical properties of carbon nanotubes (CNT) have been extensively discussed since the first studies of their optical absorption by Kataura et al. were presented in 1999 [1]. The nonlinear saturable absorption and high third-order nonlinearity make CNT-based devices firm candidates to becoming an important component of the next-generation all-optical switches [2] and passively mode-locked lasers [3]. In particular, CNT-based mode-lockers exhibit characteristics, such as ultrafast recovery time, ease of fabrication and controllable broadband operation, making them desirable over more established technologies based on semiconductor saturable absorber mirrors (SESAM) or fiber Kerr-based nonlinear devices [3].

However, the performance of CNT-based devices is hindered by the methods currently used to deposit the CNT in the optical system which is done either by spray-coating a substrate with CNT [3] or directly synthesizing a CNT film on a substrate [4]. CNT-mode lockers directly deposited on a fiber ferrule offer compact, fiber compatible solutions but are subject to (a) physical damage from the contact between the ferrules and the CNT, (b) optical power induced thermal damage, and (c) limited interaction length with the nonlinear material. Proposed solutions to tackle the shortcomings of the first proposed configurations include embedding the CNT in a host material such as a polymer [5] and exploiting the evanescent interaction between the CNT and the propagating light [6, 7]. Both approaches have contributed to alleviate the problems previously described but thus far, the fabrication of CNT-polymer waveguide structures with low losses and efficient CNT-light interaction have not been demonstrated [8]. The results from the evanescent field interaction demonstrate high-power lasing yet it is still desirable to find a configuration that can bear higher optical powers and where longer direct interaction between the optical field and the CNT is feasible [9].

In this paper, we propose a novel approach that is based on using an optofluidic device as a solution to alleviate the restrictions of current CNT-based photonic devices. The term optofluidics has recently been coined to categorize the symbiotic association between microfluidics and photonics where the microfluidic devices and systems, also known as Labon-Chip (LOC), can be fabricated, manipulated and/or interrogated by optical means. This association is not only beneficial to researchers working in microfluidic science but it also offers new alternatives and solutions to fabricate functional photonic devices [10, 11]. Here, we propose and implement for the first time to the best of our knowledge an optofluidic device that exploits the nonlinear optical properties of CNT. The proposed device is an optofluidic mode-locker consisting of a 2- $\mu$ m diameter microchannel fabricated by femtosecond laser microfabrication through the core of an optical fiber and filled with a dispersion of CNT on a solvent.

In this particular case, we employ femtosecond laser exposure combined with chemical etching to fabricate the optofluidic device in an optical fiber [12]. This microfabrication technique relies on the significantly faster rate of chemical etching of glass that has been exposed to femtosecond laser pulses compared to the etching rate of pristine glass. The method also exploits the capability of femtosecond laser inscription to fabricate three-dimensional structures in a highly controllable manner [13, 14].

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We demonstrate saturable absorption operation and mode-locked lasing in a highly integrated and compact configuration. Stability and low insertion losses are ensured by directly fabricating the saturable absorber within the optical fiber demonstrating that this is a suitable approach to solve the drawbacks associated to the spraying technique. By having the CNTs dispersed in a fluid, the efficiency of heat dissipation is increased and thus the device can withstand significantly higher optical powers. Also owing to the geometry of the device, the CNTs are less susceptible to mechanical damage since they are not in direct contact with the fiber ferrules.

#### 2. Fabrication

The fabrication of the optofluidic device was carried out by femtosecond laser inscription followed by selective chemical etching of the photo-exposed glass. The laser-matter interaction in the femtosecond laser regime is characterized by a highly deterministic, multiphoton absorption process which implies that structural modifications in the material can be confined to the region where the laser is focused permitting previously unattainable levels of control in the dimensions and properties of the inscribed structures. This allows the inscription of buried, three-dimensional structures by translating the sample with respect to the focal point of the focused femtosecond pulse [15]. Furthermore, glass modified by a femtosecond laser is etched at a much higher rate than pristine glass. This technique can be used for the fabrication of three-dimensional microfluidic devices and it does not require clean room facilities making it a fast, simple and cost-effective procedure [13, 14].

For this study, we used the same fabrication method that we first reported and applied to refractive-index sensing [12]. A femtosecond laser emitting at 800 nm was tightly focused on the fiber using a 100X objective lens with a numerical aperture of 0.55 and a working distance of 13 mm, the focus waist is estimated to be  $1.3\mu$ m. The pulse width of the laser was approximately 150 fs and the repetition rate 1 kHz with pulse energy of ~150nJ. The fiber was fixed on a dual-axis, air-bearing translation stage so that the desired structure could be written by translating the fiber with respect to the laser beam at a writing speed of  $10\mu$ m/s. As it is shown in Fig. 1(a), in order to avoid beam distortion caused by the cylindrical geometry of the fiber, a glass slip was affixed to the fiber perpendicular to the direction of propagation of the incoming laser beam, index-match oil between glass slip and fiber was employed to minimize such distortion.

Following the inscription, the fiber with the laser modified microstructure was chemically etched in a 5% HF acid solution. An ultrasonic bath was used to enhance the penetration of HF acid into the laser-modified area. The microchannels were inscribed through the core, as shown in the schematic of Fig. 1(b), and had a diameter of approximately 2  $\mu$ m and a length of 125  $\mu$ m. In this first demonstration, only a single channel was employed. However, it must be pointed out that this fabrication technique is flexible and the dimensions of the structure can be modified by adjusting parameters such as the pulse energy, writing speed, repetition rate and the focusing conditions. Hence it is possible to produce microslots with the desired length along the core, expanding the interaction length of the device [16], or adjacent to the core in order to exploit the evanescent field interaction.

For the preparation of the CNT solution, we employed commercial CNTs fabricated by the High-Pressure CO Conversion (HiPCO) method. In order to exploit efficiently the optical properties of CNTs, it is necessary to disperse the CNTs that tend to bundle together as a result of Van der Waals interactions. To avoid such agglomeration, we used the same method that is used to disperse the CNT prior to spraying of CNT in a substrate [3] which is depicted in the inset of Fig. 1(b) and consists on dispersing the CNT by ultrasonification in Dimethylformamide (DMF) solvent and then putting the solution through centrifugation. The concentration of CNT in the solution was approximately 70 parts per million (ppm) and only the visually homogeneous part of the CNT solution was used for the experiment. We worked with a low concentration of CNT to ensure minimal agglomeration. The microchannel was placed in a V-groove and immersed in a CNT solution with capillary forces contributing to the filling of the microchannel.

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Fig. 1. (a). Set-up for the fabrication of the microfluidic channel. (b) Schematic of the  $2\mu m$  microfluidic channel filled with a CNT solution. (b) Inset: Description of method to disperse CNT in DMF.

The absorption characteristics of the CNT solution in DMF used for this experiment are shown in Fig. 2(a). The absorption characteristics of the solution are only dependent on the purity of the CNTs and the efficiency of their dispersion since DMF is transparent at the relevant wavelengths. Absorption peaks at approximately 1450 nm and 1330 nm can be identified. Those peaks correspond to the allowed transitions between the Van-Hove singularities that give rise to the nonlinear saturable absorption for mode-locking [3]. The solution was then inserted into the microchannel. Figure 2(b) shows the Raman spectrum measured inside the microchannel. Several peaks can be distinguished; as well as the three sharp narrow peaks corresponding to the SiO<sub>2</sub> seen at Raman shifts of 481 cm<sup>-1</sup>, 1462 cm<sup>-1</sup>, 1526 cm<sup>-1</sup>, we can see the G and D Bands characteristic of the carbon at 1587 cm<sup>-1</sup> and 1333 cm<sup>-1</sup> respectively. The most relevant peaks are the radial breathing modes (RBMs) with Raman shifts from 200 cm<sup>-1</sup> to 290 cm<sup>-1</sup> which confirm the presence of CNTs within the microchannel.



Fig. 2. (a). Absorption spectra of the CNT dispersed in DMF, the dashed line indicates the expected non-saturable absorption [3], dotted line indicates the wavelength at which the laser here implemented operates. (b) Raman spectra from the CNT-filled microchannel indicating the presence of CNTs.

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#### 3. Results

The 2-µm diameter, CNT-filled micro-channel was inserted into a fiber laser ring cavity such as shown in Fig. 3(a). By inscribing the microchannel in a standard optical fiber, the insertion of the saturable absorber in the laser cavity is simplified, and the overall insertion losses can be minimized. The measured overall losses of the saturable absorber were less than 0.5dB. The laser gain medium used was an erbium-doped fiber amplifier (EDFA). One optical isolator built-in the EDFA and a second isolator were used to ensure unidirectional operation within the laser cavity. 10% of the intracavity lasing light was coupled out as laser output while the remaining 90% was launched back into the cavity as feedback. Cavity dispersion was optimized by adding a several meters of single-mode fiber (SMF), which made up for a total cavity length of approximately 70 m.

The pulse operation of the fiber laser is summarized in Fig. 3. Figure 3(b) shows the optical spectrum of the laser output centered at 1566 nm with a spectral bandwidth at full-width half-maximum (FWHM) of 1.1 nm. Assuming a transform limited sech<sup>2</sup> pulse waveform, this FWHM corresponds to pulse duration of 2.4ps. The vertical, dotted line in Fig. 2(a) indicates the operating wavelength of the passively mode-locked laser, hence lasing was achieved despite not operating at the wavelength where saturable absorption was at its maximum.

The autocorrelator trace and its sech<sup>2</sup> approximation are shown in Fig. 3(c) indicating that the pulse duration is 2.3ps. The time-bandwidth product of 0.31 was calculated from the spectral bandwidth of 1.1nm and the pulse duration, matching the expected time-bandwidth product of a transform limited soliton with sech<sup>2</sup>-pulse shape which is characteristic of ring-cavity laser configurations. Finally, the photo-detector measurements in Fig. 3(d) shows a pulse train with a 390 ns interval between pulses yielding a 2.56MHz repetition rate which corresponds to the fundamental repetition rate for this laser cavity length.



Fig. 3. (a). Mode-locked ring-cavity fiber laser utilizing a CNT-filled microfluidic channel. Characterization of the passively mode-locked laser based on a CNT-filled microfluidic device (b) Optical spectrum, (c) pulse train, (d) autocorrelator trace.

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One of the key advantages of this method is that high optical powers can be achieved without damaging the CNTs. We measured an output power of 13.5dBm; this corresponds to an intracavity optical power of 23.5dBm. This intracavity power is close to one order of magnitude higher than the optical damage threshold (~15dBm) reported for direct interaction with sprayed samples [6]. The increased robustness to optical damage results from efficient heat dissipation, which is attained by having the individual CNTs suspended in a liquid. Stable mode-locking operation was monitored over several hours but thorough reliability tests are yet to be carried out.

It is known that the optical nonlinearity of a system is directly proportional to the nonlinear coefficient,  $\gamma$ , the optical power, P, and to the interaction length of the nonlinear material and the optical field, L, and in this paper, we are working with a very small interaction length of approximately 2µm between the CNT and the propagating field. However, an important advantage of the proposed fabrication method is the flexibility to inscribe complex structures without significantly complicating the fabrication process. For instance, the interaction length can be easily extended to microslots by processing multiple adjoining channels, thus interaction lengths in the order of 10s or 100s of microns are accessible [16]. The position of such micro-slots can be aligned so the micro-slot crosses or is bordering the core of the fiber allowing the exploitation of a direct field interaction or evanescent field interaction length and the losses in the device. Hence the optical absorption properties can be controlled by modifying three parameters; the concentration of CNT, the dimensions of the optofluidic structure and the location of the structure with respect to the core.

This technique is also extendable to planar configurations where the integration of optical waveguides and optofluidic devices both fabricated by a femtosecond laser have already been demonstrated [17]. Hence, we anticipate that the use of optofluidic devices as a platform to exploit of the optical properties of CNT will lead to more efficient and controlled CNT-based photonic devices that can be applied not only to the implementation of passively modelocked lasers but also all-optical switching.

### 4. Conclusion

In this paper, we demonstrate, for the first time, the feasibility of employing optofluidics to exploit the nonlinear optical properties of CNT. We propose and implement a saturable absorber consisting of a  $2\mu$ m diameter microfluidic channel inscribed through the core of a conventional single-mode fiber and filled with a carbon nanotube solution in a solvent (DMF). The losses introduced by the device are less than 0.5 dB and passive mode-locking with pulse duration of 2.3 ps is demonstrated with an intracavity power of 23.5 dBm. The low losses and high intracavity power highlight the key improvement over previous reports in terms of robustness to optical damage and compactness. In addition, the simplicity of the process used to fabricate the optofluidic device based on femtosecond laser-aided chemical etching provides a clear path towards the fabrication of more complex structures that allow more efficient interaction between the CNTs and the lightwave.

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