

# Graphene-filled hollow optical fiber saturable absorber for efficient soliton fiber laser mode-locking

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**Abstract:** We demonstrate a novel in-line saturable absorber based on hollow optical fiber (HOF) filled with graphene composite for high power operation of mode-locked fiber laser. Evanescent field of guided mode propagating in few centimeter-long HOF interacts with the graphene/polyvinyl acetate (PVAc) composite, which enables robust and efficient nonlinear absorption leading to stable passive mode-locking. The mode-locked fiber laser generates soliton pulses with 5.9-nm spectral bandwidth and its maximum output power is measured up to 80 mW. We also observe passive harmonic mode-locking of soliton laser delivering stable pulses with a repetition rate of 506.9 MHz at 33rd harmonics.

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## 1. Introduction

Researches on low dimensional carbon nanomaterials such as carbon nanotubes (CNTs) and graphene have been intensively carried out in recent years for applications as future photonic and optoelectronic devices due to their unique mechanical, electrical and optical properties [1, 2]. The nonlinear saturable absorption element for passive mode-locking of the lasers [3–6] is one of the successful photonic application examples of carbon nanomaterials that possess huge optical nonlinearity with fast recovery time. Saturable absorbers (SAs) based on single-walled CNTs (SWCNTs) have been demonstrated both in fiber and bulk mode-locked lasers at various spectral ranges [3–5, 7, 8]. However, proper treatment of morphology of CNT nanostructures to control its agglomeration is required to provide efficient saturable absorption function [8]. Diameter and chirality control of CNT is also necessary for energy bandgap design depending on the operational wavelength of the lasers considered. The SA based on graphene can potentially avoid such drawbacks of CNT-SA due to its unique energy bandgap feature, i.e. point-bandgap with a linear dispersion of Dirac electrons, which enable ultra-broadband operation of graphene-SA without chirality control or bandgap engineering.

The first mode-locking of fiber laser using multilayer graphene film has been reported in 2009 [6] where few layer graphene transferred to the fiber connector ferrule was used as a SA. This approach is, however, liable to cause mechanical damage or deformation of graphene layer by direct connection between fiber ferrules and thermal damage during high power operation. The limited nonlinear interaction of few layered graphene film is not also desirable for fiber lasers generally requiring the SA with large nonlinear modulation depth. To overcome these issues, lateral interaction scheme based on evanescent wave interaction on side-polished fiber has been proposed [9] though it exhibits polarization dependent loss due to its structural asymmetry. Moreover, the laser output spectrum includes some ripples, which is mainly due to the subcavity formation during the device fabrication and irregular scattering through the surface of the side-polished fiber.

In this work, we propose a hollow optical fiber (HOF) filled with graphene/polymer composite as a novel saturable absorber. The graphene composite including polyvinyl acetate (PVAc) was employed to suppress the deformation and/or distortion of graphene morphologies [10]. Guided mode in the HOF interacts with the graphene/PVAc composite based on evanescent wave interaction over whole length (59 mm) of HOF. Our lateral interaction scheme increases damage threshold of the SA resulting in high power operation of mode-locked fiber laser. The measured maximum average output power is 80 mW at a repetition rate of 15.36 MHz. The well-shaped laser output spectrum without signature of ripple or continuous wave (CW) peak at central position is obtained with 3-dB spectral bandwidth of 5.9 nm. We also observe passive harmonic mode-locking phenomenon by adjusting polarization states of the laser cavity where the maximum repetition rate is measured as 506.9 MHz, corresponding to 33rd harmonics of fundamental mode.

## 2. Preparation of the graphene SA

The graphene is prepared as a form of reduced graphene oxide (RGO) by the solution-based modified Hummer method [11]. The chemically oxidized graphene oxide in aqueous suspension is ultrasonicated to form highly-dispersed graphene oxide and washed in deionized water multiple times. Dispersed graphene oxide is re-dispersed in DMF and PVAc is subsequently added to obtain PVAc-buffered graphene suspension. It turned out that polymer buffers could suppress the deformation of graphene sheets inside the dispersion [10]. Before injecting as-prepared graphene/PVAc solution, transmission characteristics of graphene/PVAc composite on glass plate is measured as shown in Fig. 1. The UV-VIS-NIR transmission spectrum verifies broadband absorption property of graphene over visible to IR spectral region unlike CNTs exhibiting localized absorption band. The inset of Fig. 1 is a transmission electron microscope (TEM) image, which indicates few layers morphology of graphene is well sustained in the dispersion.

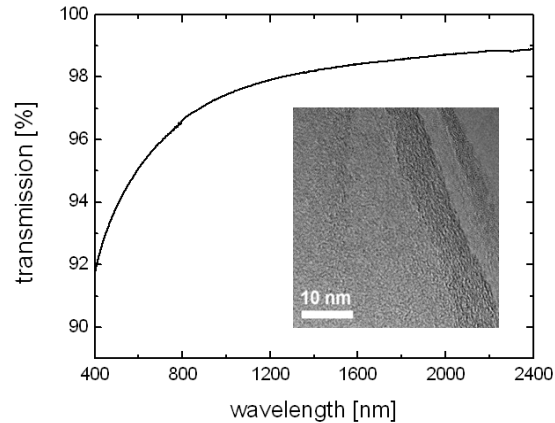


Fig. 1. Linear transmission spectrum of graphene/PVAc film on glass and transmission electron microscope (TEM) image of graphene/PVAc composite (inset).

As-prepared graphene/PVAc solution is then injected into the air hole of HOF. The HOF is composed of a central air hole,  $\text{GeO}_2\text{-SiO}_2$  ring core, and  $\text{SiO}_2$  cladding as described in Fig. 2(a), which has been originally designed for applications of optical filters or spatial mode converters [12]. When the ring-shaped core of HOF is filled with the materials with gas or liquid form, the ring-core mode of HOF can efficiently interact with the material selected [13]. In our case the graphene/PVAc solution is injected into the air-hole of HOF using a pump. The HOF used in the experiment has a ring core with 9.3- $\mu\text{m}$  outer diameter and 3.3- $\mu\text{m}$  inner-diameter. The image in Fig. 2(b) clearly indicates that the HOF is filled with graphene/PVAc solution. The sample is then fully dried at 100°C on hot plate for several hours to evaporate solvents (water and DMF) from the dispersion as shown in Fig. 2(c). This can reduce unwanted scattering losses or connection loss during splicing with normal SMF. Finally the dried graphene-injected HOF is connected with SMF using a fusion splicer under optimized condition. Thus the core-mode in the conventional SMF is adiabatically coupled to the ring-core mode in the HOF without significant loss. The ring-core mode interacts with the graphene filled over whole length of HOF as described in Fig. 2(d) and it is then coupled back to the SMF again, which acts as efficient in-line saturable absorber. In our experiment, total length of HOF used was 59 mm and the insertion loss of the sample was measured to be -1.3 dB including connection losses with normal SMFs. Our graphene SA using a HOF exhibits negligible polarization dependent loss due to its centrosymmetric structure unlike the case of side-polished fiber. The measured nonlinear saturation loss is estimated more than 2% based on modified experimental setup of Ref. 14 including optical fibers.

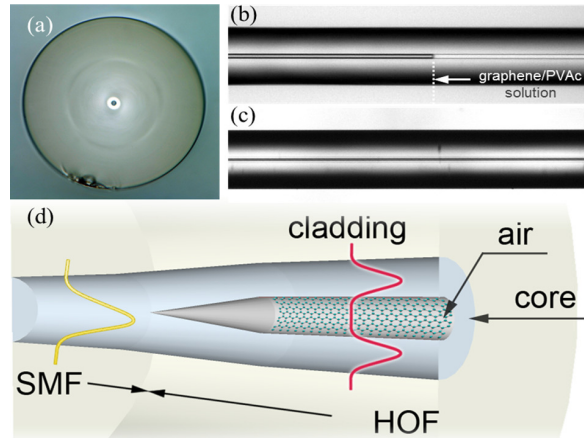


Fig. 2. (a) Microscopic image of cross-section of HOF. (b) Microscopic image of lateral cross-section of graphene/PVAc solution injected HOF and (c) fully dried graphene/PVAc injected HOF. (d) Schematic of nonlinear interaction of graphene in HOF.

### 3. Experimental results and discussions

Figure 3 shows a schematic of ring cavity fiber laser including the graphene-filled HOF-SA. The Er-doped fiber is pumped by a 980-nm laser diode through a wavelength division multiplexing (WDM) coupler. A fiber isolator is installed for unidirectional operation of laser. A polarization controller adjusts the polarization state of propagating light to match the polarization state of propagating light in each round trip and a tunable output coupler partially extracts light from the laser cavity. The fabricated-graphene SA is then inserted into the laser cavity. The inset in Fig. 3 shows the microscopic image of the splicing section between conventional SMF and HOF where adiabatic mode transition occurs through gradual collapse of the central air-hole of the ring-core in the HOF. The total dispersion of the laser cavity is estimated to be  $-0.289 \text{ ps}^2$  including the HOF possessing small anomalous dispersion of  $4.5 \text{ ps/km/nm}$  around  $1555 \text{ nm}$  [13], where soliton pulsating was expected.

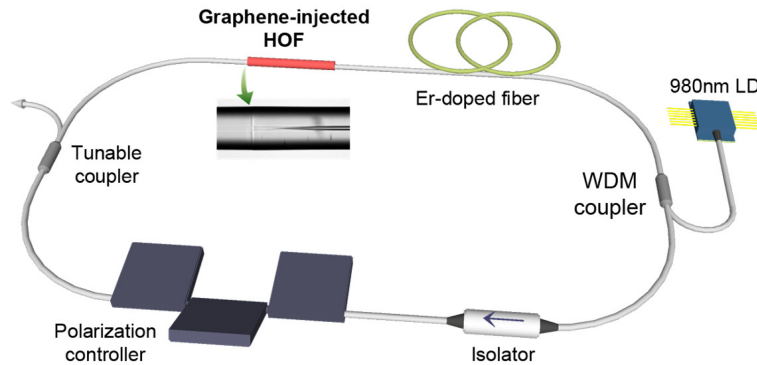


Fig. 3. Experimental setup of mode-locked Er-doped fiber laser using graphene-injected HOF. Inset figure shows the splicing point image between normal SMF and HOF.

Figure 4 represents the experimental results of the mode-locked fiber laser developed in the present work. Stable self-starting mode-locking is observed from the pump power of  $120 \text{ mW}$  where the measured laser output power and corresponding pulse energy are  $4.0 \text{ mW}$  and  $0.26 \text{ nJ}$ , respectively. The laser output spectrum and pulse duration is shown in Fig. 4(a). The central wavelength of laser output is measured to be  $1555 \text{ nm}$  with a 3-dB spectral bandwidth

of 5.9 nm. The laser output spectrum is well-defined without ripple or CW peak at central position. The pulse duration was measured by intensity autocorrelator with 2.2-m of SMF pig-tail from the laser output. The full-width half maximum of the pulse is measured to be 510 fs as shown in the inset of Fig. 4(a). Assuming generated output as soliton pulses, time-bandwidth product (TBP) is estimated to be 0.373. The difference from the value (0.315) of transform-limited pulse is mainly caused by SMF extension at laser output. Figure 4(b) and its inset represent radio-frequency (RF) spectrum and pulse train of the mode-locked fiber laser, respectively. The fundamental repetition rate was measured to be 15.36 MHz, which corresponds to the laser cavity length about 13.5 m. The main peak of fundamental repetition rate shows the extinction of  $-70$  dB from noise level at resolution bandwidth of 10 Hz, indicating stable single-pulse mode-locking.

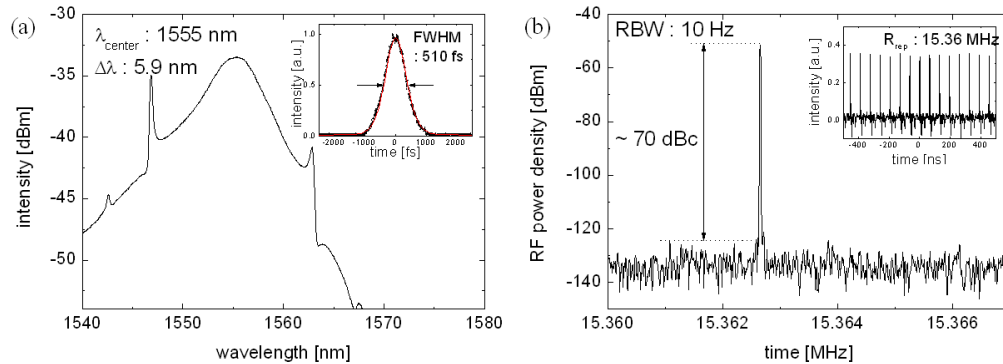


Fig. 4. (a) Optical spectrum of output pulse of mode-locked fiber laser and measured pulse duration (inset). (b) RF spectrum of mode-locked laser and pulse train (inset).

As increasing pump power up to 400 mW, we observed laser output power is increased to 80 mW at output coupling ratio of 98% while maintaining single pulse generation as shown in Fig. 5(a). The pulse energy is estimated to be 5.2 nJ in this case. It is interesting to note that single-pulse operation is switched to harmonic pulse generation at this pump power level by adjusting the polarization state of fiber at intra-cavity as shown in Fig. 5(b). Such phenomenon, called passive harmonic mode-locking, is originated from soliton energy quantization, which produce multiple fundamental soliton pulses evenly spaced in one round-trip by time-dependent gain relaxation process [15–20]. The maximum repetition rate obtained was 506.9 MHz at 33rd harmonic order. To our knowledge, this is the highest value ever reported for the graphene SA mode-locked fiber laser. The super-mode suppression ratio was measured about  $-25$  dB, which could be further improved by optimizing SA properties [21]. The background noise level is suppressed by  $-65$  dB from RF peak of 506.9 MHz at the resolution bandwidth of 1 Hz as described in the inset of Fig. 5(b). The realization of such laser with repetition rate more than GHz can be possible by optimizing the laser cavity dispersion at higher pump powers, which can be potentially useful for the applications in optical communication system and optical frequency metrology.

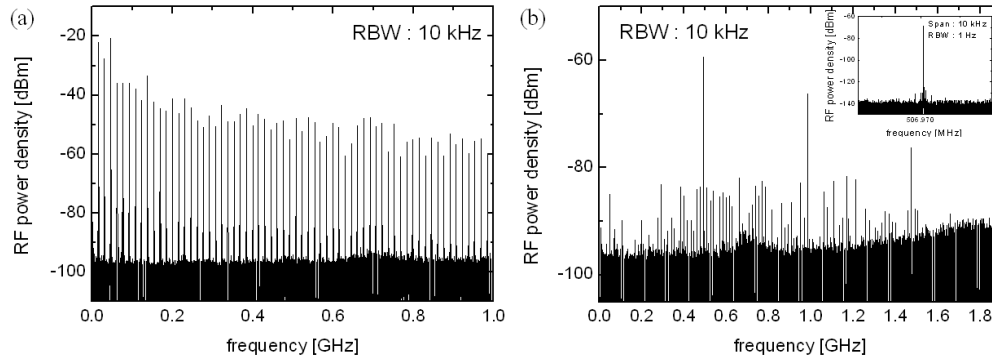


Fig. 5. (a) 1-GHz-span RF spectrum of mode-locked fiber laser with fundamental repetition rate of 15.36 MHz. (b) RF spectrum of 33rd harmonic pulse at a repetition rate of 506.9 MHz and its extended view (inset).

#### 4. Conclusion

In conclusion, we propose novel in-line graphene SA that is well suited for high-power operation of the fiber laser through robust and efficient nonlinear interaction with evanescent field in the HOF. The fabricated SA is successfully applied into fiber laser system to demonstrate stable and self-starting passive mode-locking with high output powers up to 80 mW. The fiber laser exhibit well-defined spectral shape with 3-dB spectral bandwidth of 5.9 nm and pulse duration of 510 fs at a repetition rate of 15.36 MHz. We also observe passive harmonic mode-locking of fiber laser with a repetition rate of 506.9 MHz which corresponds to 33rd harmonics of fundamental one of 15.36 MHz.

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