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Graphene Oxide-Based *Q*-Switched Erbium-Doped Fiber Laser *

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We demonstrate a pulsed ring erbium-doped fiber laser based on graphene oxide (GO), employing a simplified Hummer's method to synthesize the GO via chemical oxidation of graphite flakes at room temperature. By dipping a fiber ferrule end face onto the GO suspension, GO is successfully coated onto the end face, making it a simple saturable absorption device. A stable Q-switched pulsed fiber laser is achieved with a low pump threshold of 9.5 mW at 980 nm. The pulse repetition rate ranges from 16.0 to 57.0 kHz. The pulse width and the pulse energy are studied and discussed.

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Graphene has attracted increasing attention in photonics and optoelectronics because of its remarkable optical properties, in particular the saturable absorption. It can provide the nonlinear saturable absorption which is observed as a consequence of Pauli blocking,[1,2] and has a wide operating spectral range covering whole telecomm bands that originates from its point band gap structures.[3,4] By using graphene, various authors have realized passive mode locking[2−11] or *Q*-switching[12−16] in fiber lasers and free space solid state lasers. Nonetheless, the point band gap structure has some limitations in graphene for device applications in nanoelectronics. To solve the problem, a gap can be introduced in graphene sheets by chemical treatment. Graphene oxide (GO), an example of chemically treated graphene, has most recently emerged as a potential alternative to graphene. Absorption of oxygen on graphene opens up the band gap. GO not only enables tunable electronic, optical, and mechanical properties possible by means of controlled oxidation,[17] but also has similar ultrafast characteristics as that of graphene.[18,19]

For laser and optoelectronics applications, free-space solid state pulsed lasers using GO as a saturable absorber (SA) have been demonstrated.[17,19] In this Letter, we demonstrate a kind of novel graphene oxide absorber fabricated using pure graphite material. By using this material, inserting it into a ring laser resonator, with erbium-doped fiber (EDF) as the active gain medium, we obtain stable *Q*-switching pulses.

Many fabrication methods and synthesis processes of graphene oxide currently involve tedious steps and long experimental time, especially in mixing, heating or cooling the reactants necessary for chemical oxidation. In this work, we prepared the GO using the simplified Hummer's method as described in Ref. [20], which has the advantage of spending much shorter time in mixing the reactants. Furthermore, the whole process can be carried out without any temperature control. Oxidation of graphite flakes was carried out by mixing H_2SO_4:H_3PO_4 (320:80 mL), graphite flakes and potassium permanganate (18 g), using a magnetic stirrer. The mixture was left for stirring at room temperature for three days. Sulphuric acid was added later on to stop the oxidation process. Here graphite oxide was formed. It was washed then with 1 mol hydrochloric acid aqueous solution and repeatedly with deionized water till pH = 4–5 was obtained. The washing process was carried out using simple decantation of supernatant via centrifugation. When washed with deionized water, the graphite oxide experienced exfoliation, resulting in the thickening of the graphene solution, forming a GO gel. Figure 1 shows the graphite flakes used to synthesize the GO and the GO in deionized water.

Figure 2 shows the UV-visible absorption spectra of GO in water. The spectra are plotted in the wavelength range from 200 to 800 nm for water. An absorption band was observed at 228 nm of excitation, with absorbance intensity of briefly 2.29. This result is in

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close proximity to that of graphite oxide as reported in Ref. [21]. The strong absorption band is due to the $\pi \rightarrow \pi^*$ transitions of aromatic C = C bonds. The plot below 200 nm can be ignored, due to fluctuations in baseline measurement, which is the deionized water. A small 'shoulder' is observed at ~300 nm, possibly contributed by the $n \rightarrow \pi^*$ transitions of C = O bonds. The inset in Fig. 2 is a typical tapping mode atomic force microscopy (AFM) image of GO deposited on a freshly cleaved mica substrate.

Fig. 1. (a) Graphite flakes (3061 Insutex Industries Sdn. Bhd.) used in preparing graphene oxide. (b) Graphene oxide dispersed in deionized water.

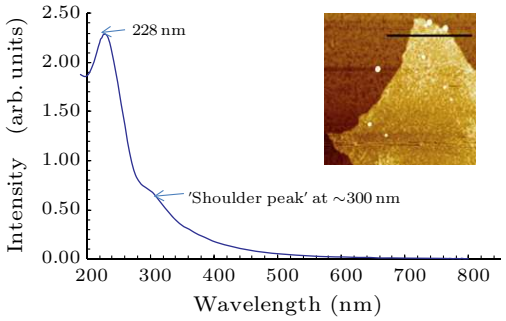

Fig. 2. UV-visible absorbance spectra of graphene oxide in water. The inset is an AFM tapping mode image of GO deposited on mica substrate. (scan area: 1 μm × 1 μm).

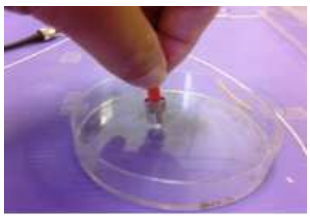

Fig. 3. A GO-based device was made by gently dipping a fiber ferrule end face onto the GO suspension.

To make a GO-based SA, we employed the 'dip-coating' method (Fig. 3), where the fiber ferrule was gently moved onto the graphene oxide solution, and the graphene oxide suspension will stick to the fiber end surface accordingly, due to considerable adhesive force. This way of transferring GO onto a fiber ferrule end to prepare a GO-based saturable absorber is done here for the first time. Compared to the method in Ref. [22], the method used here has the most exciting advantages of being simple, direct and efficient without involving tedious and long experimental procedures and time. Massive scalability is possible. The GO suspension is readily available on the fiber ferrule for connecting it with another fiber pigtail via a connector (FC/PC) to make it a simple saturable absorber device, suitable for use in Q-switching and mode-locking of fiber lasers. Further comparison with that in Ref. [23], shows this method is cost effective, i.e. it does not require costly setup and complicated tunable optical parameters. In addition, the fiber ferrule does not have the risk of being damaged by high intensity light in the core. By further controlling the concentration of the suspension during the GO synthesis process, we believe that this method is a promising candidate for low-cost and simple fabrication of the graphite-material-based saturable absorber device in an all-fiber format. The fiber ferrule was then allowed to dry at room temperature for one hour. Figure 4 shows the experimental configuration of the GO-based erbium-doped fiber laser. An isolator ensures unidirectional light propagation. The polarization controller (PC) is to maintain the polarization state of the light after each round trip propagation and thus improving output pulse stability. The cavity length is ~12.7 m.

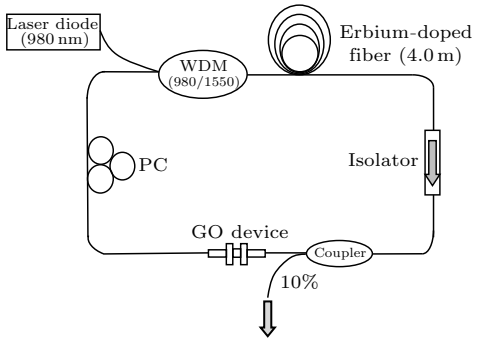

Fig. 4. GO-based erbium-doped fiber laser (EDFL) ring configuration.

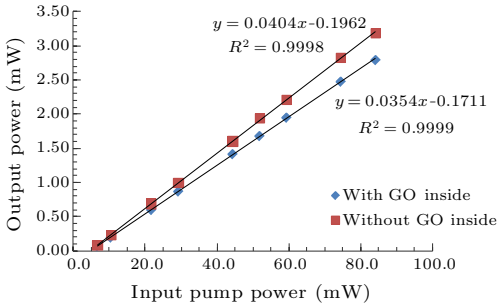

Fig. 5. Output power as a function of input power at 1558 nm.

To optimize the laser performance of pulse operation, we used the output coupler with transmittance of 10%. The lasing oscillation of typical continuous wave (cw) operation pump threshold was about 4.8 mW. When the pump power is over 9.5 mW, the laser exhibits Q-switched operation. Figure 5 shows the cw (without the graphene oxide) and Q-switched

(with the graphene oxide) operation of the average output power as a function of incident pump power. At maximum pump power (84.0 mW), the maximum output powers of the cw and Q-switched laser were 3.2 mW and 2.8 mW, respectively. The lower slope efficiency compared with the one at cw operation is due to the intracavity losses caused by the added GO for Q-switching. The ratio of the output power between GO pulsed and cw operation has a value of approximately 0.88, indicating a low insertion loss of the GO flakes, having great potential to be used as a novel saturable absorption device.

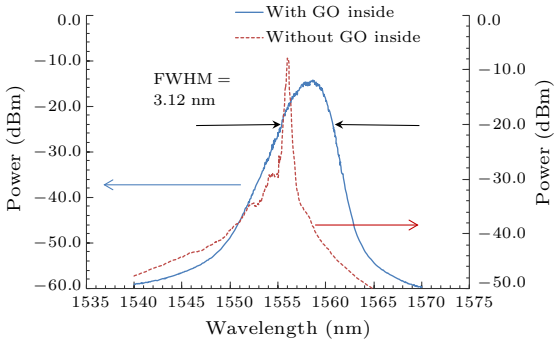

Fig. 6. Typical laser spectrum of an EDFL, with and without the GO device inside the cavity.

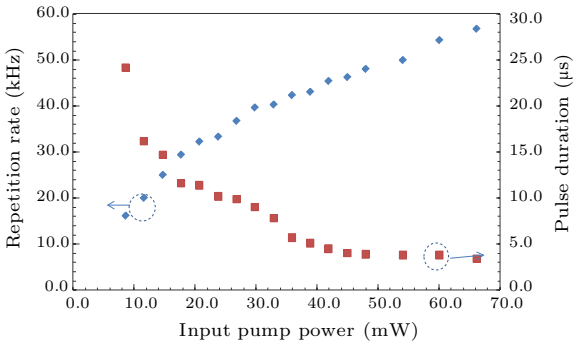

Fig. 7. Pulse width and repetition rate as a function of the incident pump power.

The cw and Q-switched pulse spectrum was measured and investigated by an optical spectrum analyzer (Anritsu MS9780A). The spectrum for both cw and pulse operation is shown in Fig. 6. The central wavelength is 1558 nm. The spectral width at the full width at half maximum (FWHM) of the pulsed operation is 3.12 nm. It can be seen that the spectral width of the pulsed operation is broader than the cw operation.

To study the characteristics of the Q-switched pulses, we observed the variation of repetition rate and pulse width by only manipulating the pump power. Figure 7 shows the relationship between the pulse repetition rate and pulse width with the pump power. The repetition rate varies from 16.0 kHz to 57.0 kHz, with a rate of 22.7 kHz for a 13.6 mW pump power. In mode-locked lasers, the cavity length determines the repetition rate.[24] This is not so in the case of Q-switched lasers, where the repetition rate is very much dependent on the pump power.[24] When the pump power is increased, more gain is available to cause saturation to the saturable absorber. As pulse generation relies on saturation, the repetition rate would have a linear relationship with the pump power, i.e. the repetition rate increases with pump power.[24] The pulse width decreases with increasing pump power, which is consistent with the findings of Ref. [12]. It is shown here that the pulse width can be significantly narrowed down from about 24.0 μs to about 3.90 μs by changing the pump power from ∼10.0 mW to ∼70.0 mW. It is believed that the pulse width could be further reduced by having a shorter length of the laser cavity design and optimizing the cavity loss.[25,26] Figure 8 shows the typical pulse train and single pulse trace at a pump power of approximately 13.6 mW.

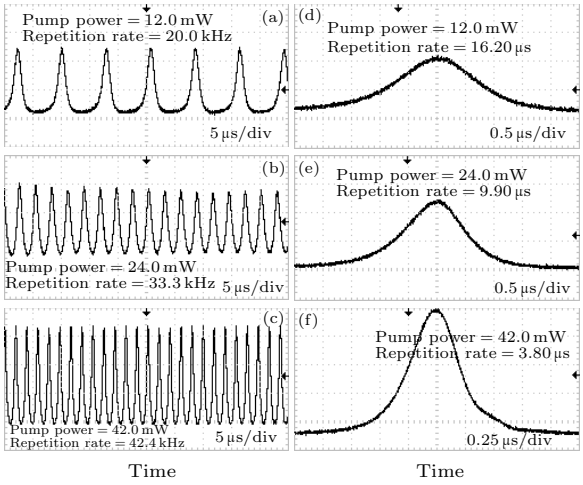

Fig. 8. Typical oscilloscope traces of Q-switched pulse train (a–c) and extended single pulse shape (d–f), showing the pulse repetition rate and pulse width respectively under different pump powers. (a) Pump power 12.0 mW, repetition rate 20.0 kHz; (b) pump power 24.0 mW, repetition rate = 33.3 kHz; (c) pump power 42.0 mW, repetition rate 42.4 kHz; (d) pump power 12.0 mW, pulse width 16.20 μs, (e) pump power 24.0 mW, pulse width 9.90 μs; (f) pump power 42.0 mW, pulse width 3.80 μs.

The single pulse energy E can be calculated using the expression

$$E = \frac{P_{\text{out}}}{\nu},$$

where P_{out} is the average output power and ν is the pulse repetition rate. Figure 9 depicts the dependence of single pulse energy against pump power. As it is shown, the single pulse energy increases monotonously along with the pump power. The largest pulse energy of 38.0 nJ is obtained at the pump power of about 70.0 mW. It is clear that the single pulse energy is linearly dependent on the pump power. The energy varies from 8 nJ to approximately 40 nJ, corresponding to the range of pump power from 10.0 mW to 70.0 mW. Larger pulse energy could be enabled by increasing

the pump power, using high gain fiber for large energy storage[27] and optimizing the coupling output and cavity loss.[25]

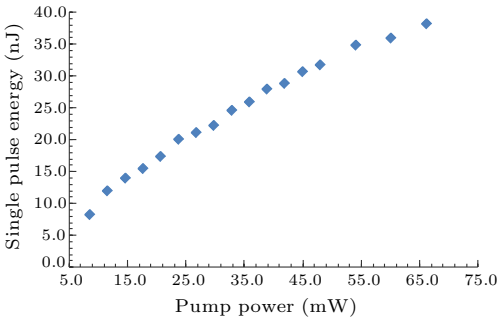

Fig. 9. Single pulse energy as a function of pump power.

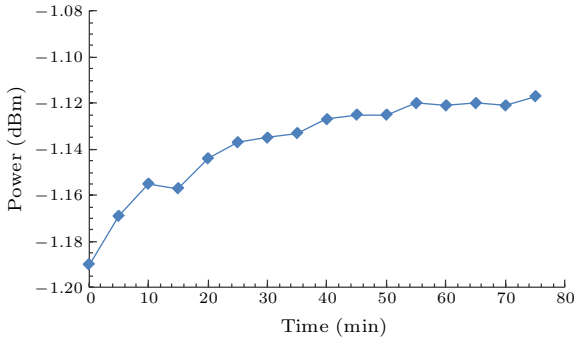

Fig. 10. Fluctuation of average power of the Q-switched pulse.

Figure 10 shows the Q-switched pulse power fluctuation. It can be seen that the average output pulse power experiences some increase against the observation time. The slight increment could be due to temperature fluctuation of the laser diode used in the experiment over time. The diode is not thermoelectrically cooled (cooler-less), hence small temperature fluctuation in the pump laser is inevitable, causing slight changes in the pump power, though the monitoring process is carried out at a constant driving current. Nonetheless, the overall power fluctuation of about 0.1 dB, corresponding to less than 3% in about 70 min, is within the measurement error and accuracy of the optical power meter used, indicating a good short-term stability of the pulsed laser output.

In conclusion, a GO based Q-switched pulsed fiber laser has been achieved. We have demonstrated Q-switching of fiber laser using graphene oxide as a saturable absorber device. The GO-based device is well fitted for passive Q-switching of fiber lasers in the 1550 nm spectral range. Under the 9.5 mW pump power from the 980 nm laser diode, pulses begin to form, with an average output power of 0.17 mW. The repetition rate increases from 16.0 to 57.0 kHz when the pump power is varied from ∼10 mW to ∼70 mW. The corresponding pulse energy is 81 nJ at 84.0 mW of pump power. The pulse has pulse width changing from 24.0 to 3.90 μs.

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