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Graphene saturable absorber for Q-switching and mode locking at 2 µm wavelength

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Abstract: Graphene saturable absorber mirror (SAM) was successfully fabricated by transferring large-size graphene flake on dielectric coating mirror. The graphene transferred on the mirror was tested by Raman spectrum measurement and scanning electron microscope imaging. With the graphene SAM, passive Q-switching and continuous wave (CW) mode locking were experimentally demonstrated in a bulk laser at 2 µm wavelength.

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References and links

1. Introduction

Graphene, a single carbon atomic layer, has attracted much attention since it was discovered in 2004 [1]. Besides its great potential in electronics, graphene has also shown its fascination in photonic applications. In 2009, Bao et al. proposed that the optical absorption in graphene could be saturated under strong excitation due to Pauli blocking, and experimentally demonstrated passive mode locking in an erbium fiber laser by using of graphene saturable absorber [2]. Further investigations show that graphene has an ultrafast recovery time and moderate modulation depth, which suggests that it is a suitable saturable absorber for passive mode locking and Q-switching. Graphene is a zero-bandgap material and has a linear dispersion, and the absorption in graphene is only determined by optical conductivity constant and independent of optical frequency. It has already been verified that graphene has an ultra-broad absorption range from visible to THz waveband [3], thus graphene is an alternative for ultra-broadband saturable absorbers.

Pulsed lasers at 2 µm wavelength have important applications in laser lidar, surgical operation, molecule spectroscopy, micro-machining of semiconductor, generation of X ray near “water window” waveband, etc [4–6]. Q-switching and mode-locking are the general techniques to generate pulsed laser. For passive Q-switching and mode locking operation, generally a saturable absorber is required to initiate and sustain the pulsing operation against CW operation in a laser. At present, semiconductor saturable absorber mirrors (SESAM) were widely used as mode-locking and Q-switching elements in 1-1.5 µm waveband. Unfortunately, the fabrication technique of SESAM was not well developed at 2 µm waveband. Due to ultra-broadband saturable absorption, graphene thus becomes an excellent alternative for 2 µm saturable absorbers.

At present, studies on passive Q-switching and mode locking with graphene as saturable absorber are mainly focused on 1-1.5 µm waveband [7–16]. However, there are few reports on 2 µm Q-switched and mode locked lasers based on graphene [17,18].

In this paper, we report on passive Q-switching and mode locking performances of a bulk laser at 2µm wavelength with graphene as saturable absorber. The graphene SAM was successfully fabricated by transferring the graphene flake onto dielectric mirror, and large-area graphene was grown by chemical vapour deposition process. The graphene SAM was tested by Raman spectrum measurement and scanning electron microscope imaging, showing graphene on mirror had a good quality.
2. Fabrication and test of graphene SAM

The large-area graphene film was grown on a copper foil with chemical vapor deposition (CVD) process by mixing CH\textsubscript{4} and H\textsubscript{2} gases at 1000 °C [19]. Firstly, the polymethylmethacrylate (PMMA) in chlorobenzene was spin-coated on the as-grown graphene film, then the copper foil was etched away by Marble’s reagent solution. After the copper foil was completely etched away, the PMMA-supporting graphene film was washed with deionized water, then transferred onto a dielectric coating mirror. The fabrication of graphene SAM was accomplished after PMMA was dissolved by acetone. The Raman spectrum and scanning electron microscope (SEM) image of graphene SAM were shown in Fig. 1. In the Raman spectrum, the G peak and 2D peak of graphene could be clearly observed. The G peak locates at ~1590.6 cm\textsuperscript{-1} with a FWHM of ~18 cm\textsuperscript{-1}, and the 2D peak is at ~2649.8 cm\textsuperscript{-1} with a FWHM of ~40 cm\textsuperscript{-1}, respectively. The intensity ratio of G peak to 2D peak is about 0.418. According to [20,21], the thickness of graphene film on mirror should be 1-2 atomic layers. Due to the intense Raman signal of mirror substrate below 1500 cm\textsuperscript{-1}, the D peak of graphene was submerged in the background signal. The graphene SAM was then tested by scanning electron microscope (SEM) imaging, an imaging region of ~200 × 200 \(\mu\)m\textsuperscript{2} was shown in Fig. 1(b). From the image of SEM, the graphene film on mirror was clear, continuous and uniform across the imaging region, showing a good quality.

![Fig. 1. (a) Raman spectrum of Graphene SAM; (b) scanning electron microscope (SEM) image of graphene SAM.](image)

3. Laser setup

The schematic of the laser setup is shown in Fig. 2. A single-emitter laser diode at 790 nm was used as the pump source. Through two convex lens, the pump light was focused into crystal with a spot size of 26 × 86 \(\mu\)m. The Tm:CLNGG crystal employed in the experiment had a length of 9 mm and Tm-doping concentration of 3 at.% in melt. Tm:CLNGG was a disordered crystal with a broad gain linewidth [22]. The crystal was placed with Brewster angle for minimizing the transmission loss for p polarization light. For effectively removing the heat while pumping, the crystal was wrapped with indium foil and tightly mounted in a water-cooled copper block. For quasi-three-level Tm-ion laser, water cooling is vital for efficient laser operation. In the experiment the circulating water temperature was set at 9.0 °C. An X-folded cavity was employed to obtain a suitable laser mode size in the crystal and on the graphene SAM. By ABCD matrix method, the laser mode radius was calculated to be ~35 \(\mu\)m in the crystal and ~40 \(\mu\)m on the graphene SAM. The concave mirrors M1, M2, and M3 have a same radius of curvature of 10 cm, and were highly reflectively coated at laser wavelength and anti-reflectively coated at pump wavelength. The wedged output coupler had a transmission of 2% at 2000 nm wavelength. In the experiment, a slit was inserted in the cavity for suppressing the high-order transverse mode oscillation. A pair of CaF\textsubscript{2} prisms with a tip to tip distance of 40 cm was employed for intracavity dispersion compensation. The as-
fabricated graphene SAM was used as passive Q-switching and mode-locking elements in the laser. The reflectivity of graphene SAM was measured to be about 95% under low incident light intensity and the modulation depth was ~1% at 2 µm wavelength.

Fig. 2. The schematic of the laser setup, LD: laser diode; F1, F2: convex lenses; M1, M2, M3: concave mirrors with same radius of curvature (ROC) of 10 cm; OC: output coupler; graphene SAM: graphene saturable absorber mirror.

4. Experimental results and discussions

With graphene as the saturable absorber, the stable Q-switching operation could be obtained in the laser. The typical Q-switched pulse trains are shown in Fig. 3. The Q-switched pulses have a long pulse duration of 9 µs, which is attributed to the low modulation depth of the 1-2 layer graphene and long laser cavity used in the experiment. According to the Fig. 3(b), the Q-switched pulses have a repetition rate of 5.8 kHz. In this case the average output power of the laser was 40 mW, thus the corresponding Q-switched pulse energy was 6.9 µJ. The Q-switched pulses were very stable and pulse to pulse intensity fluctuation was estimated to be less than 5%.
However, when we increased the incident pump power, damage of the graphene SAM was observed in the experiment. With a larger laser mode size on the graphene SAM, we believe the average output power of the Q-switched laser could be further increased. The laser spectrum of the Q-switched laser is shown in Fig. 4. The laser spectrum is centered at 2013.5 nm, with a narrow bandwidth of only ~1.2 nm.

Fig. 4. Spectrum of the Q-switched pulses.

By carefully optimizing the laser spot position on the graphene SAM and aligning the laser cavity, stable CW mode locking could be achieved in the laser. A maximum average output power of 60 mW was obtained in the mode locked laser. In the experiment, the mode locking was monitored for nearly one hour and it remained stable. When further increasing the pump power, damage on graphene SAM was found. The mode locked pulse trains were shown in Fig. 5. The mode-locked pulses had a repetition rate of 95 MHz, corresponding to the laser cavity length of 1.58 m. By checking the pulse trains in different time scales, no Q-switching envelope was observed, and the laser operated in CW mode locking regime. In the laser with graphene as saturable absorber, the laser could operate in Q-switching and CW mode locking regime under the same pump power, which could be attributed to monolayer and bilayer graphenes existing on graphene SAM. When the intracavity laser was incident on bilayer graphene, the modulation depth and non-saturable absorption loss were larger, thus the laser operated in Q-switching regime with a low average output power. While the intracavity laser was incident on monolayer graphene, the modulation depth and non-saturable absorption loss were less, thus CW mode-locking could be initiated and higher average output power could be obtained. For graphene SAM, various graphene layers can be deposited at different positions on a mirror, which will bring convenience for optimizing Q-switching and mode locking.
performances of lasers. In order to evidence the function of graphene SAM as saturable absorber in the laser, we translated the graphene SAM in the transverse direction perpendicular to cavity axis. When the laser spot was moved out from graphene region, Q-switching and mode locking signals vanished.

With a commercialized autocorrelator, the autocorrelation trace of the mode locked pulses was measured, as shown in Fig. 6. Assuming a Sech² shape, the mode-locked pulse duration was 882 fs. The spectrum of the mode locked pulses was measured by using an optical spectrum analyzer with a resolution of 0.22 nm. The laser spectrum is centered at 2014.4 nm, with a bandwidth of 6.1 nm. The time-bandwidth product is calculated to be 0.39, which is close to Fourier transform limit value for Sech²-shape pulses.

5. Conclusion

In conclusion, a graphene SAM was successfully fabricated and the passively Q-switched and mode locked laser based on the graphene SAM was experimentally demonstrated at 2 µm wavelength. The Q-switched laser generated microsecond pulses with pulse energy of 6.9 µJ, and the CW mode-locked laser emitted pulses with pulse duration of 882 fs, repetition rate of 95 MHz and average output power of 60 mW. The experimental results show that the graphene SAM is a promising saturable absorber for passive Q-switching and mode locking at 2 µm wavelength.

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