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# Graphene mode-locked femtosecond laser at 2 $\mu$m wavelength

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We experimentally demonstrated a passively mode-locked femtosecond laser by using a graphene-based saturable absorber mirror (graphene SAM) in the spectral region of 2 μm. The graphene SAM was fabricated by transferring chemical-vapor-deposited, high-quality, and large-area graphene on a highly reflective plane mirror. Stable mode-locked laser pulses as short as 729 fs were obtained with a repetition rate of 98.7 MHz and an average output power of 60.2 mW at 2018 nm. 

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Graphene, a single-atom thin sheet of carbon atoms with a honeycomb lattice, has attracted much attention due to its unique electronic and photonic properties [1]. The Pauli blocking of electron states makes graphene usable as a saturable absorber for passive mode locking [2]. Moreover, graphene has the intrinsic advantages of ultrafast recovery time [3], lower saturation energy fluence [2], and easy fabrication. Since graphene mode locking has been demonstrated in Er-doped fiber lasers [2,4], a series of fiber and bulk lasers have been reported that use graphene as a saturable absorber for ultrashort pulse generation [5–11]. Graphene has zero bandgap and a linear dispersion relation, so, theoretically, it could be used as a saturable absorber for mode locking over an ultrawide spectral range from the visible to far-infrared [2,5]. However, so far, almost all of the reports on graphene mode-locked lasers have been focused on the spectral region of 1 to 1.5 $\mu$m. At 2 $\mu$m wavelength, only Liu *et al.* reported on a passively mode-locked Tm:$YAlO_3$ laser by using graphene oxide as a saturable absorber, which generated 10 ps pulses [12].

Ultrafast lasers operating in the 2 $\mu$m spectral region have important applications in ultrafast molecular spectroscopy, remote sensing, optical communications, and mid-infrared laser generation by an optical parametric process. Up to now, most of the 2 $\mu$m passively mode-locked lasers were realized based on semiconductor saturable absorber mirrors (SESAMs) [13–17]. Compared with a graphene saturable absorber, a SESAM generally has a narrower operation bandwidth (~tens of nm) and requires very complex fabrication processes. Specifically, a SESAM is wavelength-relative and requires the bandgap of a semiconductor material that matches with the photonic energy, which limits the operation wavelength range for the SESAM. On the other hand, graphene Q-switch demonstrated that graphene could be used as a saturable absorber at the 2 $\mu$m region [18].

To date, graphene saturable absorbers used for mode-locking was generally fabricated by graphene—polymer composites [4,5], graphene sheets exfoliated from graphite in the liquid phase [11], mechanical exfoliation [19], and CVD [2,10]. In contrast to other processes, the CVD method could synthesize high-quality and large-area graphene with the required layers, which was desirable for mode locking.

In this Letter, we demonstrated a passively mode-locked femtosecond laser at 2 $\mu$m wavelength by using a graphene SAM, which was fabricated by transferring CVD-deposited high-quality and large-area graphene on a broadband, dielectric-coated mirror. Stable mode-locked pulses as short as 729 fs were obtained with an average output power of 60.2 mW and a repetition rate of 99 MHz at 2018 nm. To the best of our knowledge, this is the first femtosecond operation in the 2 $\mu$m spectral region with graphene as a saturable absorber.

The graphene film was grown on a 25 $\mu$m thick Cu foil using CVD by mixing $CH_4$ and $H_2$ gases at 1000 °C [20]. The graphene film was spin-coated with 5 at.% polymethylmethacrylate (PMMA) in chlorobenzene after growth, then the Cu substrate was etched away by Marble's reagent solution ($CuSO_4$:HCl:$H_2O$ = 10 g:50 ml:50 ml). The PMMA-supported graphene was washed with de-ionized water and then transferred onto a highly reflective plane mirror. The fabrication of the graphene SAM was accomplished after PMMA was dissolved by acetone. Figure 1 shows the image of the graphene SAM (inset) and the Raman spectrum of graphene excited by a 514.5 nm laser source. The transferred graphene layer on the highly reflective mirror had a large size of ~1.3 cm × 1.3 cm, as shown in the inset. The reflectivity of the graphene SAM around 2 $\mu$m was measured to be about 95%. The G-band peak in the Raman spectrum was located at ~1581 $cm^{-1}$ with a FWHM of about 25.0 $cm^{-1}$, and the 2D-band peak was at ~2685 $cm^{-1}$ with a FWHM of about 37.6 $cm^{-1}$. The intensity ratio of the G-band peak to 2D-band peak was about 0.26. According to a previous analysis [21,22], the thickness of graphene on the mirror should be 1 to 2 layers. The weak D-band peak of graphene could be observed at ~1347 $cm^{-1}$ after subtracting the Raman signal of a highly reflective mirror substrate. The weak D-band peak suggests few defects in graphene. Otherwise, an optical microscope image of the graphene

 

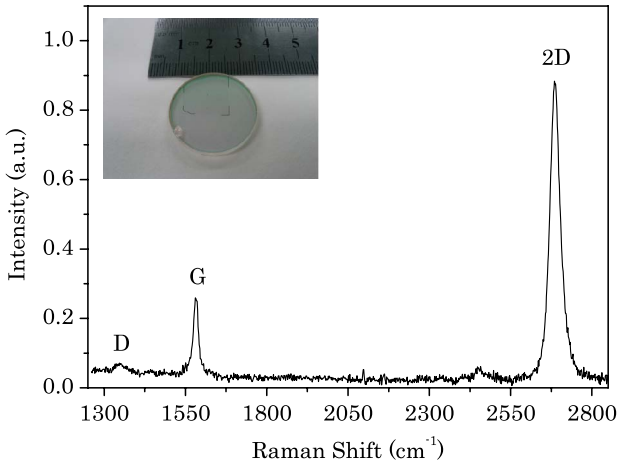


Fig. 1. (Color online) The Raman spectrum of graphene excited at 514.5 nm. The Raman signal of the mirror substrate was subtracted. Inset shows the image of the graphene SAM used in the experiment.

SAM with 200× magnification was shown in Fig. 2. The image reflected a real graphene area of ~1.0 mm× 0.9 mm, which was much larger than the laser mode size on graphene SAM (~tens of $\mu$m). In Fig. 2(a), the graphene boundary could be clearly identified. In Fig. 2(b), the graphene layer was clean, continuous, and uniform across the whole region, showing a good quality of the graphene SAM.

The schematic of the mode-locked laser setup based on the graphene SAM is shown in Fig. 3. A Brewster-cut, 9 mm long Tm-doped calcium lithium niobium gallium garnet (Tm:CLNGG) crystal with $Tm^{3+}$ concentration of 3 at.% in melt was used as the gain media. The crystal was wrapped with indium foil and tightly mounted in a water-cooled copper block with the cooling water temperature set at 9.0 °C. The pump source was a commercial single-emitter AlGaAs laser diode at 790 nm (nLight Laser, NL-C-5.0-790-3-F). The two coupling lenses of F1 and F2 have the same focal length of 100 mm. In the experiment, a standard X-folded cavity was used for achieving a suitable laser mode size in the crystal (~35 $\mu$m radius) and on the graphene SAM (~40 $\mu$m radius). A slit was settled close to the output coupler (T = 2%) to suppress the high-order transverse mode oscillation in the vertical direction. To obtain a femtosecond operation regime, a pair of $CaF_2$ prisms with a tip-to-tip distance of 39 cm were inserted in the laser resonator to compensate for the cavity dispersion.

By optimizing the position of the graphene SAM and adjusting the laser cavity carefully, stable continuous

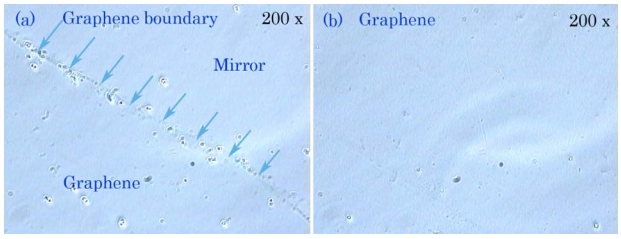


Fig. 2. (Color online) Image of a graphene film transferred to the highly reflective mirror under an optical microscope.

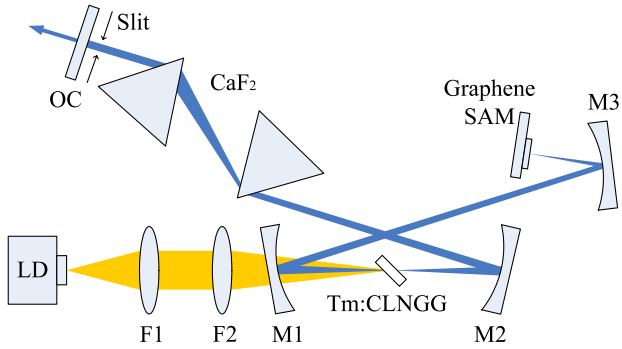


Fig. 3. (Color online) Experimental setup of the mode-locked laser based on the graphene SAM. The focus length of F1 and F2 was 100 mm. M1, M2, and M3 have the same radius of curvature of −100 mm. OC: output coupler.

wave (CW) mode locking could be obtained. Beyond 1.21 W of absorbed pump power, the CW mode locking was self-started in the laser. The threshold of CW mode locking was only 38 mW of average output power, corresponding to intracavity power of about 1.90 W. Compared with the SESAM mode-locked laser [23], the CW mode-locking threshold in the laser was much lower, which could be attributed to the lower saturation energy fluence and the modulation depth of the few layers of graphene. The modulation depth of the graphene SAM was estimated to be ~1%. In the experiment, we monitored the mode locking for nearly an hour, and it remained stable. The typical CW mode-locked pulse trains in nanosecond and millisecond time scales are shown in Fig. 4. The pulse repetition rate was 98.7 MHz, corresponding to the laser cavity length of 1.52 m. The maximum average output power was 60.2 mW under the absorbed power of 1.58 W. The output laser had a round $TEM_{00}$ mode. If we further increased the pump power, the graphene SAM was easily damaged in the experiment.

The mode-locked pulse's duration was measured by a commercial autocorrelator (APE, PulseCheck 50). The autocorrelation trace and optical spectrum are shown in Fig. 5. The autocorrelation trace has a pulse duration of 729 fs FWHM, assuming a $sech^2$-shaped pulse. The spectrum of the laser is centered at 2018 nm with a FWHM bandwidth of 7.3 nm, which was measured by

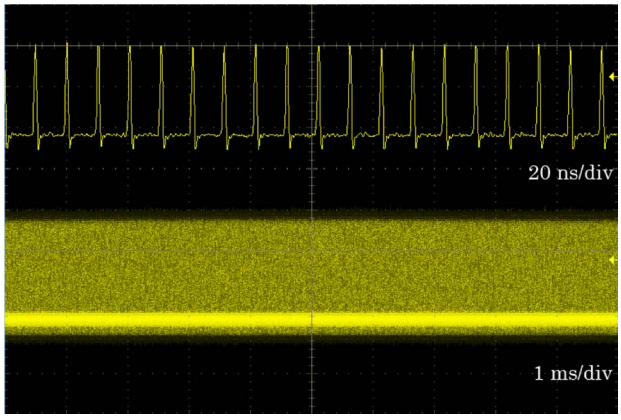


Fig. 4. (Color online) CW mode-locked pulse trains in nanosecond and millisecond time scales.

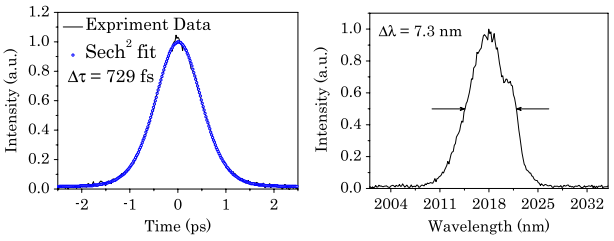


Fig. 5. (Color online) (Left) Autocorrelation trace and (right) optical spectrum of the mode-locked pulses.

a mid-infrared optical spectrum analyzer with a resolution of 0.22 nm. The time–bandwidth product of the mode-locked pulses is calculated to be about 0.39, which is close to the Fourier transform limit value for the $\mathrm{sech}^2$-shaped pulses.

In conclusion, we have experimentally demonstrated a diode-pumped passively mode-locked femtosecond laser at a 2 $\mu$m wavelength by using a graphene saturable absorber mirror. The laser-generated mode-locked pulses had a pulse duration as short as 729 fs, a repetition rate of 99 MHz, and an average output power of 60.2 mW at 2018 nm wavelength. To the best of our knowledge, this is the first demonstration of a graphene mode-locked femtosecond laser at 2 $\mu$m wavelength. The experimental results suggest that graphene is an excellent saturable absorber for femtosecond pulse generation in the 2 $\mu$m spectral region.

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