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Graphene mode-locked femtosecond laser at 2 μm 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. © 2012 Optical Society of America

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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 μm. At 2 μm wavelength, only Liu et al. reported on a passively mode-locked Tm: YALO3 laser by using graphene oxide as a saturable absorber, which generated 10 ps pulses [12].

Ultrafast lasers operating in the 2 μm 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 μm 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 μm 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 μm 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 μm spectral region with graphene as a saturable absorber.

The graphene film was grown on a 25 μm thick Cu foil using CVD by mixing CH4 and H2 gases at 1000 °C [20]. The graphene film was spin-coated with 5 at.% poly-methylmethacrylate (PMMA) in chlorobenzene after growth, then the Cu substrate was etched away by Marble’s reagent solution (CuSO4·HCl·H2O = 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 μm 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...
SAM with 200× magnification was shown in Fig. 2. The image reflected a real graphene area of \( \sim 1.0 \text{ mm} \times 0.9 \text{ mm} \), which was much larger than the laser mode size on graphene SAM (\( \sim \) tens of \( \mu \text{m} \)). 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. M1, M2, and M3 have the same radius of curvature of \( -100 \text{ mm} \). OC: output coupler.

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
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 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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