Monolayer graphene saturable absorbers with strongly enhanced evanescent-field interaction for ultrafast fiber laser mode-locking

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Abstract: We demonstrate an efficient all-fiber saturable absorber (SA) that evanescently interacts with a graphene monolayer. Strong nonlinear interaction between the graphene sheet and evanescent wave was realized in both experiments and numerical calculations by employing an over-cladding structure on high-quality monolayer graphene that uniformly covered the side-polished fiber. A passively mode-locked Er-doped fiber laser was built, including our in-line graphene SA, which stably generated ultrashort pulses with pulse duration of 377 fs at a repetition rate of 37.7 MHz. The corresponding 3-dB spectral bandwidth of the laser was measured to be 8.6 nm at the central wavelength of 1607.7 nm. We also experimentally observed that the spectral bandwidth and pulse duration of the laser output could be controlled by proper selection of the refractive index of the over-cladding material on the monolayer-graphene SA.

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1. Introduction
Graphene, a two-dimensional atomic layer of carbon atoms forming a hexagonal lattice, has been regarded as a promising material for use as a building block for future photonic, optoelectronic, and plasmonic devices owing to its unique electrical and optical properties [1–3]. In particular, its outstanding features including large Kerr nonlinearity, ultrafast carrier dynamics, and ease of integration into diverse optical systems enabled the graphene to apply to novel nonlinear optic systems such as wavelength-independent nonlinear signal generation [4] and broadband nonlinear saturable absorption for passive mode-locking of lasers. After the first demonstration of mode locking of fiber lasers based on multilayer graphene [5], there have been a number of research efforts on fiber mode-locked lasers based on graphene saturable absorbers (SAs) in various spectral ranges with diverse configurations [6–15]. In the majority of these works, the SA was fabricated by transferring the layered graphene [5–10] or graphene flake/polymer composite [11–15] onto the optical fiber connector ferrules. In spite of its simplicity in fabrication, a fiber-ferrule–based SA obviously exhibits a weak optical absorption in the thin graphene layer because of limited nonlinear interaction length. Moreover, mechanical damage by direct physical contact or optically induced thermal damage to the graphene layer can potentially restrict the performance of the fiber laser, particularly at high power. Any parasitic reflection at a fiber connector included in the laser cavity also may deteriorate the performance of mode-locked lasers.

In order to avoid the drawbacks described above, an indirect interaction scheme achieved through an evanescent-field interaction has been suggested based on several different platforms. For example, a coating of graphene oxide (GO) on a tapered fiber [16] or side-polished fiber (SPF) [17] was proposed as an in-line SA. Multilayer graphene/polymer composite film transferred to an SPF [18] was also reported for the nonlinear interaction of the graphene with the evanescent field. A hollow-core fiber filled with reduced graphene oxide (RGO) solution [19] and a photonic crystal fiber injected with graphene nanoparticles [20] were also used as in-line SAs for graphene–evanescent-field interaction. This lateral interaction scheme guarantees a long nonlinear interaction length and reduced optical power in the interaction region, potentially providing a large modulation depth and a high optical damage threshold to the graphene-based SAs. However, the irregular structure of graphene
flakes in the RGO or GO solution can lead to inevitable scattering loss, resulting in large non-saturable loss in the SA. Insufficient nonlinear interaction with graphene materials also causes limited performance of fiber lasers both in spectral and temporal properties [17]. Although the SA using the evanescent-field interaction of uniform monolayer graphene film potentially reduces this unnecessary non-saturable loss, an efficient mode-locker using evanescent-field interaction with a monolayer graphene has yet to be realized because of its weak nonlinear interaction.

In this paper, we demonstrate an efficient in-line graphene SA that evanescently interacts with a uniform graphene monolayer for stable fiber-laser mode locking. We experimentally observed that there is strong interaction with the evanescent field in the presence of an index-matched over-cladding layer on the uniform graphene monolayer covered with the SPF. The polarization-dependent optical absorption properties of the graphene layer on the SPF were experimentally explored as a function of the refractive index of the over-cladding medium, and the results agree reasonably well with our numerical expectations. A passively mode-locked Er-doped fiber laser was built based on our in-line graphene SA; the resulting laser stably delivers solitonic pulses with a pulse duration of 377 fs and a spectral bandwidth of 8.6 nm at a central wavelength of 1607.7 nm. We also observed that the pulse duration and the spectral bandwidth of the laser output could be adjusted by changing the refractive index of the over-cladding medium that covered the graphene monolayer on the SPF.

2. Design, fabrication, and characterization of graphene SAs

We numerically investigate the electric-field distribution of the guided mode in an SPF covered with a graphene monolayer to design efficient graphene SAs. Figures 1(a) and (b) compare the numerical results calculated with air and index-matched \((n = 1.444)\) over-claddings, respectively. In the calculation, we set the minimum distance between the fiber core–cladding boundary and the polished surface to approximately 0.5 μm, based on a consideration of the fabrication conditions in the experiment. In Fig. 1(a), the electric-field distribution of the fundamental guided mode is off-center from the fiber core because of the air over-cladding where the field intensity in the graphene layer was estimated as 0.4% of the maximum intensity of the guided mode. In the presence of the index-matched over-cladding, it can be clearly seen that the evanescent field is further extended to the graphene layer, allowing for a strongly enhanced graphene–light interaction. Figure 1(c) summarizes the calculation results as a function of the over-cladding index. As the over-cladding index approaches the cladding index of the optical fiber, the intensity in the graphene layer greatly increases. For example, in the case of an over-cladding index of 1.444, the relative intensity ratio \(I_{\text{at graphene}} / I_{\text{max}}\) was estimated to be 23.6%, which is about an enhancement of a factor of 60 compared to that with an air over-cladding; this will enable strong nonlinear interaction between monolayer graphene and the evanescent field.
Figure 2(a) illustrates the fabricated monolayer graphene SA on an SPF. A conventional single-mode fiber (SMF 28e) was buried in a quartz block and side-polished until the index-oil drop loss became – 45 dB. Monolayer graphene was grown on a copper foil using a thermal chemical vapor deposition (CVD) method [21]. As a supporting layer, a poly(methyl methacrylate) (PMMA) film was spin-coated on the graphene layer with a thickness of 250 nm. After etching the copper foil using an ammonium persulfate solution, the graphene/PMMA film was transferred to an SPF, and the PMMA film was then dissolved by acetone. Figure 2(b) shows a microscopic image of the graphene monolayer without PMMA film on the SPF, which indicates that large-area (7x5 mm²) monolayer graphene is uniformly transferred to the SPF section. The interaction length of light with graphene was estimated to be 3 mm in our SPF. We carried out the micro-Raman measurement of our graphene sample at several points, and Fig. 2(c) shows a typical Raman spectrum result. We observed that our monolayer graphene film has a nearly uniform Raman intensity ratio between the 2D peak and the G peak \(I_{2D}/I_G = 4.69\) with a well-suppressed defect peak, indicating the high quality of the fabricated graphene sample over a large area. We then examine the linear optical transmission properties of the fabricated graphene SA by using several different index oils as over-claddings. Figure 2(d) summarizes the experimental results, which are compared with the results of numerical calculations. In our SA, the propagating light selectively couples with the graphene film for different input polarizations owing to the asymmetric geometry of the SA. We experimentally observed that the graphene layer predominantly interacts with the transverse electric (TE) mode whose polarization direction is parallel to the graphene surface, resulting in polarization-dependent loss (PDL) of the device. In the experiment, the PDL of the graphene SA increases from 0.09 dB to 13.1 dB as the over-cladding index varies from that of air to that of fiber cladding. As shown in Fig. 2(d), the experimental results agree reasonably well with the numerical calculations when the complex refractive index of the graphene is approximately regarded as 3.22 + 2.62i, in accordance with Ref. 22.
The nonlinear transmission properties of our graphene SA were investigated by using a lab-built mode-locked fiber laser along with a motorized variable attenuator and an optical power meter. A pulse laser with a pulse duration of 400 fs was used as an input source, where the incident optical power and the polarization state were adjusted by a computer-controlled variable optical attenuator and a polarization controller, respectively. The optical transmission was simultaneously monitored by the optical power meter through a computer. Figures 3(a) and (b) present the experimental results for the SA with air and index-matched over-claddings, respectively. The graphene SA with the air over-cladding initially exhibits a small insertion loss of about 6.7% at low power levels; this insertion loss includes both scattering losses and optical absorption by the graphene layer. The optical transmission gradually increases with increasing input power, resulting in a 0.2% modulation depth with a saturation fluence of 75.6 MW/cm². In the case of the graphene SA with index-matched over-cladding, the device initially exhibits a large optical absorption of about 95.2% in a monolayer graphene film. Although we observed that the optical transmission increased by more than 5%, as shown in Fig. 3(b), the absorption did not reach the full saturation level because of the power limit of currently available our pulsed laser source.

3. Fiber-laser experiment

A ring-cavity Er-doped fiber laser was built as schematically depicted in Fig. 4. A hybrid component serving as an optical isolator, a wavelength division multiplexing (WDM) coupler, and an output coupler that extracts 10% of the light from the laser cavity were employed to simplify the laser cavity setup. A heavily Er-doped fiber (EDF) was used as a gain medium, which is optically pumped by a 976-nm laser diode (LD). The polarization state in the laser cavity was adjusted with a polarization controller (PC), and a fabricated in-line graphene SA with an index-matched over-cladding was inserted into the laser cavity for mode locking. The total cavity length of the fiber laser was measured to be 5.49 m, where the estimated total cavity dispersion was – 0.11 ps².

Figure 5(a) shows the optical spectrum of the laser output, where the measured average output power was 5.41 mW at an applied pump power of 120 mW. The typical spectral shape of an optical soliton was observed, where the measured spectral bandwidth was 8.6 nm at the
central wavelength of 1607.7 nm. Assuming a soliton pulse, the pulse duration measured by an intensity autocorrelator was 377 fs, which fits well with the soliton pulse shape as shown in Fig. 5(b). The fundamental repetition rate was measured to be 37.72 MHz from the radio frequency (RF) spectrum shown in Fig. 5(c), which is in agreement with the laser cavity length of 5.49 m. The background noise is well suppressed with a suppression of 66.6 dB with respect to the main peak signal. The RF spectrum of the fundamental repetition rate and its beat note when viewed over a wide frequency range (inset of Fig. 5(c)) indicate stable operation of the mode-locked laser. We observed that our laser exhibits self-started mode-locking operation, which stably maintained against external perturbation of laser cavity or increase of the pump power. We further explore laser output characteristics for the SAs with different over-cladding indices. Figure 5(d) briefly summarizes the results for the spectral bandwidth and pulse duration of the laser output for a given laser cavity. We observed that the pulse duration of the laser output became shorter as the over-cladding index increased, where the measured pulse widths were 429, 395, and 377 fs for the over-cladding indices of 1.426, 1.434, and 1.444, respectively. The corresponding spectral bandwidth changes from 7.7 nm to 8.4 nm to 8.6 nm. From the result in Fig. 5(d), we expect that the SA possesses different pulsating abilities depending on the over-cladding index, which affects the pulse formation for a given laser cavity. As a consequence, this may allow one to control the spectral bandwidth and pulse duration of the laser output by proper selection of the refractive index of the over-cladding material.

Fig. 5. (a) Spectral and (b) temporal properties of the fabricated soliton-laser output. (c) RF spectrum of the laser output pulse train (inset: RF spectrum viewed over a wide frequency range) and (d) characteristics of the spectral bandwidth and pulse duration of the laser output as functions of the over-cladding index of the SA.

4. Conclusion

In summary, we report an efficient monolayer-graphene in-line saturable absorber that interacts with the evanescent field of the guided mode in a side-polished fiber. The results obtained through experiments and numerical study show that a strong optical absorption of more than 90% can be achieved without significant scattering loss in a monolayer-graphene saturable absorber by employing an index-matched over-cladding structure. The fabricated Er-doped mode-locked fiber laser including our in-line monolayer-graphene saturable
absorber stably delivers ultrashort pulses with durations of 377 fs and 3-dB spectral bandwidths of 8.6 nm. We also observed that the laser output properties including the spectral bandwidth and pulse width could be tuned by adjusting the index of the over-cladding that covered the uniform monolayer graphene. We believe that our finding of strongly enhanced interaction between monolayer graphene and the light will be helpful to realize an efficient graphene saturable absorber possessing low scattering loss and large modulation depth as well as highly efficient optoelectronic devices based on graphene.

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