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Passively Mode-locked Solid-state Laser with Absorption Tunable Graphene Saturable Absorber Mirror

Yiran Wang, Baitao Zhang, He Yang, Jia Hou, Xiancui Su, Zhipei Sun, and Jingliang He

Abstract—Two-dimensional layered materials have attracted huge interest in the generation of ultrafast laser for their excellent saturable absorption properties. However, it is still challenging to precisely control their saturable absorption properties. Here, by alternatively changing the electric field intensity on the surface of high-reflection mirror, we successfully control the nonlinear absorption properties (e.g., saturable fluence, modulation depth) of graphene-based saturable absorber mirrors (GSAM) at the optical telecommunication wavelength of 1.3 µm and their applications in solid-state lasers for the first time. Modulation depth of 1.2% is obtained from a GSAM with deposition of a $\lambda/8$ $(\lambda = 1.3 \mu m)$ thick SiO₂ layer between the monolayer graphene and a high-reflection mirror, while modulation depth is increased to 4.3% with a $\lambda/4$ thick SiO₂ layer insertion in another GSAM. Pulses with the duration of 20 ps ($\lambda/8$ thick SiO₂ insertion) and 7.4 ps ($\lambda/4$ thick SiO₂ insertion) are achieved respectively based on the two mirrors. Our results indicate that this method is easy and reliable to versatility modulate the saturable absorption properties of other two-dimensional layered materials beyond graphene for the generation of ultrafast solid-state lasers.

Index Terms—Mode-locked lasers, Graphene saturable absorber.

I. INTRODUCTION

Owing to the unique electronic band and physical properties, graphene has attracted extensive attention [1,2]. The fast carrier relaxation time, large nonlinearity and broadband optical absorption property enable graphene to work as a saturable absorber (SA) for generation of ultrashort pulses in a broad spectral range, covering from 0.8 to 2.8 µm [3-28]. In recent years, many other nanomaterials (e.g. transition-metal dichalcogenides (TMDs) [29-31], black phosphorous [32-34], etc.) beyond graphene have been demonstrated as promising SAs for ultrafast pulse generation. Compared to these recently

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emerged nanomaterials, graphene owns broader absorption bandwidth, faster recovery time and easier fabrication processes, which enable ultrafast pulse generation in a wider range [35]. Various fabrication methods of graphene have been studied, such as mechanical exfoliation [1], liquid-phase exfoliation [36], and chemical vapor deposition (CVD) [36]. Compared to other fabrication methods, the CVD approach possesses the advantages of large-area production with low defects and controlled number of layers [36], thus is more suitable for practical applications, such as mode-locked solidstate lasers. Indeed, in the past several years, CVD-grown graphene has been used in various wavelength Q-switched and mode-locked lasers [14-23, 37]. However, in those reports, the unsaturated absorption and modulation depth of graphene were difficult to control precisely compared to SESAMs.

Until now, several methods have been developed to control the absorption of monolayer graphene, such as chemical doping, electrical gating, etc. However, those methods are still limited by their drawbacks. For example, the doping concentration in graphene cannot be precisely controlled in the high concentration range with these methods [11,38,39], and the electrical gating is complicated for the additional contactpattern process [40]. In 2013, C. A. Zaugg et al. [41] have alternatively modulated the graphene's absorption by coating an insulating layer between a distributed Bragg reflectors (DBR) and a graphene flake. Using this device, a broadband (ranging from 935 to 981 nm) mode-locked vertical-externalcavity surface-emitting laser was realized. In this work, we detailed study the mode-locking performance using our designed graphene-SiO₂-integrated specially saturable absorption mirrors at 1.3 µm in solid-state lasers for the first time. 1.3 µm is an eye-safe wavelength and corresponds to the low-dispersion and low-loss spectrum of silica fibers in the telecommunication band. And mode-locked laser has a

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potential to be used for high data rate coherent communication. Using a $\lambda/4$ thick SiO₂ GSAM, pulses as short as 7.4 ps are achieved with an average output power of 0.22 W. However, with a $\lambda/8$ thick SiO₂ GSAM, pulses as short as 20 ps are achieved with an average output power of 0.75 W. To the best of our knowledge, this is the first demonstration of 1.3 µm mode-locked solid-state lasers based on absorption modulated monolayer graphene. The results illustrate that this method is reliable and flexible to adjust the absorption and modulation depth of GSAMs in solid-state lasers.

II. GSAM FABRICATION AND CHARACTERIZATION

In order to get the field intensity enhancement on the highreflection (HR) film surface, an extra SiO₂ film is coated on the surface of a HR mirror (HR @ 1.3 µm) by plasma enhanced chemical vapor deposition (Oxford Instruments PECVD 80+). The thickness of the SiO₂ film is designed as follows to tune the graphene absorption. After, a 7×7 mm² monolayer CVD graphene is transferred onto the HR mirror. The schematic image of the GSAM device is shown in the inset of Fig. 1. To examine the defects and the layer number of graphene on the GSAM, Raman measurement is performed with a laser excitation wavelength at 532 nm. Typical Raman spectrum is shown in Fig. 1. The weak D-peak intensity at 1350 cm⁻¹ shows that graphene has negligible defects. The intensity ratio (0.3) between G-peak (1589 cm⁻¹) and 2D-peak (2680 cm⁻¹) confirms a monolayer graphene film on the HR mirror [42].



Fig. 1. Raman spectrum of a typical GSAM. Inset: the image of the GSAM.

In a general standing wave cavity formed by the incident and reflected waves, the field intensity enhancement $\xi(z)$ at the distance z from the mirror can be written as [38, 43]:

$$\xi(z) = \frac{|E_{in}(z) + E_{out}(z)|^2}{|E_{in}(z)|^2}$$
(1)

where $E_{in}(z)$ and $E_{out}(z)$ are the incident and reflected wave electric fields. And the field intensity enhancement at the graphene on our GSAM can be written as [41]:

$$\xi_{abs}(d) \approx \frac{4}{1 + n^2 \cot^2(\frac{2\pi}{\lambda}nd)}$$
(2)

where n is the refractive index of SiO₂, λ is the wavelength and d is the thickness of the SiO₂ film.



Fig. 2. Electric field distribution of (a) graphene with a $\lambda/8 \operatorname{SiO}_2$ layer on the HR mirror, (b) graphene with a $\lambda/4 \operatorname{SiO}_2$ layer on the HR mirror. The green curve represents the standing electric field intensity. The black vertical line represents the position of graphene.

In our experiment, the HR mirror is designed as HR at 1.3 μ m and anti-reflection (AR) at 0.8 μ m and 1.0 μ m. Fig. 2 (a) shows the construction of our GSAM and the corresponding field intensity distribution. In our design, two GSAMs are S/(1.2H1.2L)(HL)14(0.9H0.52L)/A coated by and $S/(1.2H1.2L)(HL)^{14}(0.9H1.03L)/A$ structure, where S. H. L. and A represent substrate, high refractive index material (ZrO₂), low refractive index material (SiO₂) and air, respectively. 1.2H means 1.2 times the thickness of ZrO₂, which is defined similarly for 1.2L, 0.9H, 1.03L, and 0.52L. According to Eq. (2), the field intensity enhancement is calculated to be 1.21 at the surface of the $\lambda/8$ SiO₂ GSAM. The absorption of graphene on this HR mirror is calculated to $\xi_{abs} \times 2.3\% = 2.78\%$. However, for the $\lambda/4$ SiO₂ GSAM, the field intensity enhancement at the surface is 3.975, resulting in stronger graphene absorption, which is calculated to be $\xi_{abs} \times 2.3\% =$ 9.14%. Furthermore, the absorption could be reduced by reducing the thickness of the SiO₂ layer down to ~ 0%. Note to mention that this method can also be applied for the absorption modulation of other 2D materials at different operation wavelengths.

Fig. 3(a) shows the linear reflection spectrum of these two GSAMs between 1.3 μ m and 1.4 μ m. The linear absorption of the two GSAMs is measured to be 2.5 % and 8.2%, respectively, in good agreement with the theoretical estimation (2.78% and 9.14% for these two GSAMs). The nonlinear absorption properties are also characterized using a picosecond 1342 nm laser (Fig. 3(b)). Using the fitting formula in Ref. [44], the saturation fluence, modulation depth and nonsaturable loss of the GSAM with λ /8-SiO₂ layer are estimated to be 71 μ J/cm², 1.2% and 1.1%, respectively. However, for the device with a λ /4-SiO₂ film coating, the saturation fluence, modulation depth

and nonsaturable loss are estimated to be $15 \ \mu$ J/cm², 4.3% and 4%. Compared to the GSAM device with the λ /8-SiO₂ thin film, the saturation fluence decreases, but the modulation depth and nonsaturable loss increase because of the increased field enhancement factor. In our experiment, the highest modulation depth of 4.3 % is obtained. It is difficult to further increase the modulation depth unless the nonsaturable loss could be reduced, which can be achieved by further optimization of device design or fabrication (e.g., graphene quality).



Fig. 3. (a) Linear reflection spectrum and (b) nonlinear absorption of two GSAMs.

III. MODE-LOCKING OPERATION

As shown in Fig. 4, a Z-fold cavity is designed for the modelocking operation. In this setup, a 808-nm fiber-coupled laser diode with a core diameter of 200 µm is used as the pump source. A 3 mm × 3 mm × 5 mm Nd:YVO₄ crystal with 0.3 at. % Nd³⁺ is employed as the gain medium. During the experiment, the crystal is wrapped with an indium foil and mounted in a water-cooled copper block holder maintained at 17 °C. The pump laser is focused into the gain crystal with a beam radius of 80 µm. The dichroic mirrors M1, M3, M5 (R = ∞), M2, M4 (R = 0.5 m) and M6 (R = 0.2 m) are all AR coated at 0.8 µm and HR coated at 1.3 µm. A plano mirror with the transmission of 3% at 1.3 µm is used for output coupler (OC). With ABCD matrix propagation theory, the laser mode radii are calculated to be ~ 93 µm in the laser crystal and ~ 49 µm on the GSAM, respectively.



Fig. 4. Experimental setup of the mode-locked laser with GSAM. M1, M3, M5: plano input mirror; M2, M4: concave mirror with radius of curvature (ROC) = 500 mm; M6: concave mirror with radius of curvature (ROC) = 200 mm; OC: output coupler with 3% transmittance; X: Nd:YVO₄ crystal; LD: laser diode.

Fig. 5 shows the average output power versus the pump power with a HR mirror and two different GSAMs for performance comparison. By inserting the GSAM containing the $\lambda/8$ SiO₂ layer into the resonator, the laser turns into a stable continuous-wave mode-locking (CWML) at the absorbed pump power of 2.95 W. When the absorbed pump power increases to 4.1 W, the maximum output power of 748 mW is obtained, corresponding to a slope efficiency of 21%. When the GSAM with the $\lambda/4$ SiO₂ layer employed in the resonator, CWML operation is observed with the absorbed pump power of 2.3 W. The maximum output power of 220 mW is obtained at the absorbed pump power of ~ 2.95 W, corresponding to a slope efficiency of 11.8%. When the absorbed pump power exceeded CWML range for both two GSAM, the laser was found to run into metastable QML regime. The lower slope efficiency is caused by the larger absorption and nonsaturable loss of the GSAM. Compared with the GSAM with the $\lambda/4$ SiO₂ film, the GSAM with the $\lambda/8$ SiO₂ film is more suitable for high power mode-locked laser or low emission cross section gain medium (such as Yb doped crystals), benefitting from its relative lower insertion loss.



Fig. 5. The average output power of CWML regime versus absorbed pump power.

The mode-locked pulse duration is measured by a noncollinear autocorrelator (Pulse Check 150, APE). As shown in Fig. 6 (a), pulses as short as 7.4 ps are achieved with the GSAM containing the $\lambda/4$ SiO₂ film under the output power of 0.22 W. The corresponding spectrum with a FWHM of 0.3 nm is shown in the Fig. 6(b). The time-bandwidth product is calculated to be 0.369, which is 1.17 times larger the transformlimited value (0.315) for a sech²-shaped pulse. On the other hand, the pulse duration of 20 ps is achieved with the GSAM containing the $\lambda/8$ SiO₂ film with a corresponding FWHM of 0.16 nm. The pulse width is smaller by using the GSAM with the $\lambda/4$ SiO₂ film because of its higher modulation depth [45], which implies that our demonstrated GSAM fabrication method is useful for narrowing the pulse width. And the pulse width with GSAM containing the $\lambda/4$ SiO₂ is also smaller than other reports of graphene based 1.3 µm mode-locked laser due to the higher modulation depth [46]. The CWML pulse trains with the $\lambda/4$ SiO₂ GSAM were recorded with the period of 22.4 ns, as shown in Fig. 7 (a) with nanosecond and millisecond time span. To study the stability of the mode-locked operation, the radio frequency (RF) spectrum was measured. As illustrated in Fig. 7(b), the RF spectrum of the CWML laser with the GSAM containing the $\lambda/4$ SiO₂ film illustrates the signal-to-noise ratio of ~ 60 dB. The inset of Fig. 7(b) shows the RF spectrum

recorded on the 1-GHz span. There is no significant spectral modulation, which implies good stability performance of the mode-locked laser. We measured the long-term stability for both GSAM for half an hour, and the recorded output power showed an instability of about $\pm 2\%$.



Fig. 6. (a) The normalized autocorrelation trace with sech² fitting and (b) the corresponding output spectrum.



Fig. 7. (a) Pulse trains and (b) radio frequency spectrum of the CWML laser. The inset figure shows the 1 GHz-wide.

IV. CONCLUSION

In conclusion, the absorption of monolayer graphene is tuned by coating different thicknesses of SiO₂ films between the graphene layer and HR mirror. In this work, two GSAMs with a $\lambda/8$ thick SiO₂ layer and $\lambda/4$ thick SiO₂ layer insertion between the monolayer graphene and HR mirror are used to compare their SA performance on the generation of ultrafast laser. The highest modulation depth of 4.3% and the shorted pulse with of 7.4 ps are obtained based on the GSAM with the $\lambda/4$ thick SiO₂ layer coating. Because of the lower insertion loss, the GSAM with the $\lambda/8$ SiO₂ film is more suitable for high power modelocked laser or low emission cross section gain medium (such as Yb doped crystal), while the GSAM with the $\lambda/4$ SiO₂ film is more suitable for short pulse generation with broad spectral width. Our results indicate that this method is easy and flexible to modulate the saturable absorption properties of other 2D nanomaterials beyond graphene for the realization of ultrafast lasers.

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