Graphene-Bi_2Te_3 Heterostructure as Broadband Saturable Absorber for Ultra-Short Pulse Generation in Er-Doped and Yb-Doped Fiber Lasers

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***Abstract*—A new type of broadband saturable absorber made of graphene-Bi_2Te_3 heterostructure was reported, which synergistically combines light–matter interaction in graphene with that in a small bandgap semiconductor material Bi_2Te_3 to achieve an improved broadband nonlinear optical response. The graphene-Bi_2Te_3 heterostructure films were grown by chemical vapor deposition with 15% Bi_2Te_3 coverage on graphene, in which most of the Bi_2Te_3 nanoplatelets are less than 30 nm thick. It is interesting to find that the heterostructure thin film shows broadband saturable absorption property. At the communication band (around 1560 nm), the saturable intensity and modulation depth are measured to be 4.95 MW/cm^2 and 18.98%, respectively. While around 1067 nm, the corresponding saturable intensity and modulation depth are experimentally measured to be 2.61 MW/cm^2 and 23.11%, respectively. By incorporating this optical saturable absorber inside either an Er-doped or Yb-doped fiber laser, we are able to generate ultra-short pulse with very stable operation at 1565.6 and 1049.1 nm. Our experimental results clearly demonstrate that the graphene-Bi_2Te_3 heterostructure can be a promising broadband nonlinear optical material for broadband ultra-fast laser photonics.**

***Index Terms*—Graphene-Bi_2Te_3 heterostructure, broadband saturable absorbers, ultra-short pulse, fiber lasers.**

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I. Introduction

GRAPHENE, one-atom-thick planar sheet consisting of sp^2-bonded carbon atoms stacked in a two-dimensional (2-D) honey comb lattice, have arouse strong research interest in ultra-fast laser photonics, since the demonstration of atomic-layer graphene based optical saturable absorber (SA) [1], [2]. Because of its unique optical properties, such as low scattering loss induced by its planar morphology, wavelength-independent absorption [3], [4], ultra-fast relaxation time [5] and absorption bleaching [6], [7] at high optical intensity, graphene is considered as a remarkable SA for ultrafast pulse generation. Different operation regimes, either mode-locking or Q-switching, by graphene-based SAs have been demonstrated on either fiber lasers [8]–[15] or solid state lasers [16]–[20] from the visible towards the mid-infrared. Furthermore, wavelength-tunable operation [21], [22] and multi-wavelength operation [23], [24] had also been reported in fiber lasers. Sotor *et al.* demonstrated an all-fiber thulium (Tm-) and erbium (Er-) doped fiber laser simultaneously mode-locked by a common broadband graphene SA, allowing for the mode-locking of two lasers spectrally separated by almost 400 nm [25].

Recently, topological insulators (TIs) (such as Bi_2Se_3, Bi_2Te_3, and Sb_2Te_3) [26], [27] and transition metal dichalcogenides (such as MoS_2 and WS_2) [28] had been applied as new types of 2-D materials for optoelectronic devices. Bi_2Se_3 and MoS_2 fabricated by high-yield liquid-phase exfoliation technique show interesting nonlinear optical properties [29]–[32] and they can be fabricated as SA devices with improved (or at least comparable) nonlinear optical properties than that of graphene, such as modulation depth and saturation intensity. Zhao *et al.* presented the passively mode-locked erbium-doped fiber laser by using TI SA in 2012 [33]. Soon after that, Du *et al.* published the first report on the mode-locking operation of an ytterbium-doped fiber laser based on MoS_2 SA [34]. For the purpose of broadband operation, both TI and MoS_2 SAs have been used to mode-lock Yb-doped [35]–[37], Er-doped [38]–[41] and Tm-doped [42]–[44] fiber lasers operating at 1.06, 1.56 and 2.0 μm, respectively. Wavelength-tunable optical pulse was also obtained from the passively mode-locked fiber laser with the introduction of TI and MoS_2 SAs [45], [46].

From point of view of energy structure, zero or small bandgap materials are the most suitable candidate for broadband operation of those laser applications as they own wavelength-insensitive optical response at telecommunication bands. In particular, Dirac materials such as graphene and TIs are important broadband materials which have been applied for photonics such as photodetection [47] and pulse lasers [48]. However, there are some optical limitations in terms of absorption strength, spectral range and carrier dynamics if these 2-D materials were used individually. For instance, some shortcomings of graphene SA and TI SA need to be overcome in order to further improve the performance, such as the limited light absorption of graphene ($\pi\alpha = 2.3\%$ for monolayer graphene) [3] and heavily populated intrinsic defects of TIs which makes its transport properties dominated by the bulk state instead of the desired massless Dirac surface states [49]. Very recently, graphene-TI heterostructures, with high-quality ultrathin nanoplates of TIs (Bi_2Se_3) [50], [51] and enhanced absorption strength as well as broadened responsive spectral range [47], have been demonstrated as another interesting material for optoelectronic devices that could operate from the visible to the near-infrared range. Furthermore, Mu *et al.* have reported the generation of ultra-short pulse in Er-doped fiber laser mode-locked by graphene-Bi_2Te_3 heterostructure saturable absorber (G-Bi_2Te_3-SA) [48]. Nevertheless, the capability of this heterostructure material for broadband operation in pulse laser has not been explored. Herein, we demonstrate that graphene-Bi_2Te_3 heterostructure can be used as a broadband SA and further investigate its applications for the mode-locked Er-doped and Yb-doped fiber lasers. Based on the balanced twin-detector measurement system, its saturable absorption properties at different wavelengths have been investigated. As a result, passively mode-locked pulses with pulse duration of 1.1 ps at 1565.6 nm and 144.3 ps at 1049 nm have been achieved at mode-locked Er-doped and Yb-doped fiber lasers, respectively.

II. Experimental Results

A. Material Preparation and Characterization

The graphene-Bi_2Te_3 heterostructure SA was prepared by a similar approach as previous reports [47], [48]. First of all, monolayer graphene thin film was grown on copper substrate by chemical vapor deposition (CVD). Following, the Bi_2Te_3 nanoplatelets were grown on top of graphene film by CVD, in which graphene functions as an atomic template for the nucleation and growth of Bi_2Te_3 as these two materials share similar hexagonal lattice. By precisely control the growth parameters especially the growth time in the second round growth, thin Bi_2Te_3 single crystals with only a few quintuple layers are produced on graphene to form Van der Waals heterostructure. Fig. 1(a) shows the atomic force microscopy (AFM) image of the graphene-Bi_2Te_3 heterostructure sample with a coverage of 15% Bi_2Te_3 on graphene. The thickness of the Bi_2Te_3 nanoplatelets is less than 13 nm, as shown in the Fig. 1(b). The linear optical response of the graphene-Bi_2Te_3 heterostructure has been measured by UV-visible-infrared spectrometer in the spectral range from visible to near-infrared wavelength, as shown in Fig. 1(c). It is found that the graphene-Bi_2Te_3 heterostructure sample also has a relatively flat absorption curve from 900 to 2000 nm. The linear absorption coefficient of graphene-Bi_2Te_3 heterostructure sample is 10.15% at 1050 nm and 9.36% at 1560 nm. Finally, the graphene-Bi_2Te_3 heterostructure film is transferred onto the facet of the fiber ferrule so as to be easily integrated into fiber laser system.

Besides the linear optical response, the optical SA response of the graphene-Bi_2Te_3 heterostructure film has been investigated by using the balanced twin-detector measurement system, as shown in Fig. 2(a). A 50:50 coupler was used to equally separate the incident light into two different laser beams (A and B). Beam A is detected by detector A as the reference power (P_r) while beam B, after passing through the graphene-Bi_2Te_3 heterostructure film, is measured by detector B as the signal power (P_s). With the continuous tuning of the optical attenuator, different optical transmission with respect to different incident laser powers can be obtained, and therefore the transmission is calculated by P_s/P_r at different optical powers. Fig. 2(b) shows the saturable absorption of the graphene-Bi_2Te_3 heterostructure film at the telecommunication band. The incident optical pulse (repetition rate of 100.8 MHz, central wavelength range of 1559.5 nm, pulse width of 0.585 ps, and the maximum output power up to 16 mW) was emitted from a home-made ultra-short laser source, as shown in the insert of Fig. 2(b). Upon fitting the relation between the optical transmittance and optical powers by using the conventional

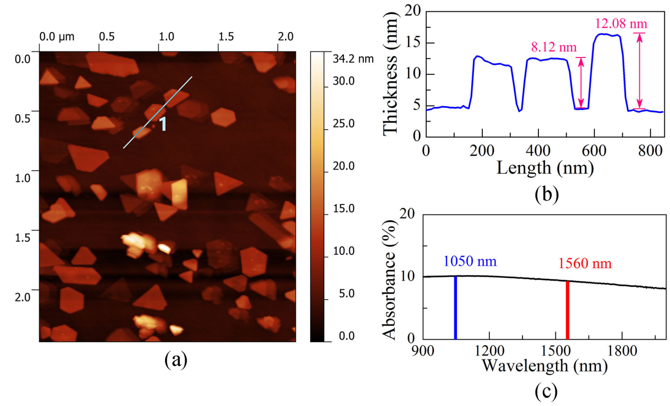

Fig. 1. (a) The AFM image and (b) thickness profiles along solid line 1 in (a) of the graphene-Bi_2Te_3 heterostructure film on SiO_2 substrate. (c) The linear optical response of the graphene-Bi_2Te_3 heterostructure.

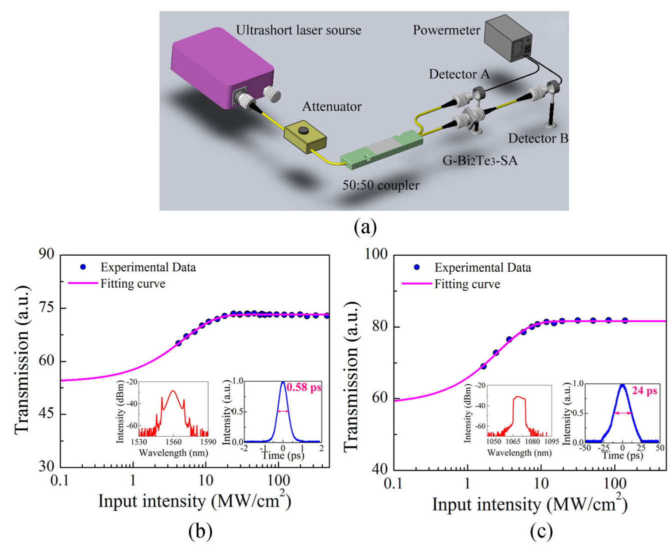

Fig. 2. (a) The experimental setup of the balanced twin-detector measurement system. G-Bi_2Te_3-SA: graphene-Bi_2Te_3 heterostructure saturable absorber. (b) The SA curves of the graphene-Bi_2Te_3 heterostructure film at 1560 nm and (c) at 1067 nm. The inserts of (b) and (c) show the spectra and autocorrelation traces of corresponding ultrashort pulse for measuring the SA curves.

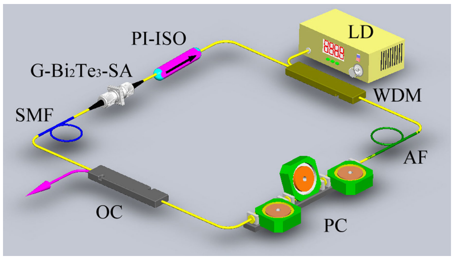

Fig. 3. Experimental setup of mode-locked fiber laser. PI-ISO: polarization independent isolator. LD: laser diode. WDM: wavelength division multiplexer. AF: active fiber. PC: polarization controller. OC: optical coupler. SMF: single-mode fiber. G-Bi_2Te_3-SA: graphene-Bi_2Te_3 heterostructure saturable absorber.

formula of $T(I) = 1 - \Delta T \exp(-I/I_s) - T_{ns}$ (where $T(I)$ is the transmission rate, ΔT is the modulation depth, I is the input intensity, I_s is the saturating intensity, and T_{ns} is the non-saturable absorbance), the saturating intensity and modulation depth are measured to be 4.95 MW/cm^2 and 18.98%, respectively. Fig. 2(c) shows the saturable absorption of the graphene-Bi_2Te_3 heterostructure film at nearly 1 μm with the input optical pulse of with repetition rate of 11.2 MHz, central wavelength range of 1067 nm, pulse width of 24.14 ps, and output power up to 20 mW, as shown in the insert of Fig. 2(c). The saturating intensity of 2.61 MW/cm^2 and modulation depth of 23.11% are been obtained after fitting, shown in Fig. 2(c).

B. *Experimental Results on Mode-Locked Fiber Laser*

In order to experimentally take advantage of the broadband saturable absorption of the G-Bi_2Te_3-SA, we had designed the experimental setup of the mode-locked fiber laser based on G-Bi_2Te_3-SA in Fig. 3. In the mode-locked Er-doped fiber laser, a piece of 3 m Er-doped fiber (LIEKKI Er16-8/125) as the active fiber with group velocity dispersion (GVD) of -13 ps^2/km is pumped by 980 nm laser diode with a 980/1550 nm wavelength division multiplexer (WDM). The polarization controller (PC) is used to slightly tune the intra-cavity birefringence. The polarization independent isolator (PI-ISO) enforces the unidirectional propagation of the intracavity optical pulse. The G-Bi_2Te_3-SA is introduced into the laser cavity as a passive mode-locker. The total cavity length is 28.95 m consisting of single mode fiber (SMF) with GVD parameter -23 ps^2/km. The total cavity GVD is -0.636 ps^2. The 10% optical coupler (OC) is used to tap the optical pulse signal. While in the Yb-doped fiber laser, the active fiber is a piece of 0.75 m long Yb-doped fiber (LIEKKI Yb1200-4/125) with a GVD of 24.22 ps^2/km pumped by 980 nm laser diode using a 980/1064 nm WDM. The PC, PI-ISO and OC with the operation wavelength at 1060 nm are also spliced into the laser cavity. The passive mode-locker is the same G-Bi_2Te_3-SA used in Er-doped fiber laser. The total cavity length is 54.18 m with a piece of 40 m long SMF with the GVD of 21.91 ps^2/km at 1060 nm, which is used to optimize mode-locking. The total cavity GVD is 1.189 ps^2. Both the optical pulses generated in Er-doped and Yb-doped fiber laser are analyzed by an optical spectrum analyzer (Ando AQ-6317B), and a 4 GHz MHz oscilloscope (DS09404A).

Due to the saturable absorption of the graphene-Bi_2Te_3 heterostructure, the self-starting mode-locking operation of the Er-doped fiber laser based on G-Bi_2Te_3-SA is obtained when

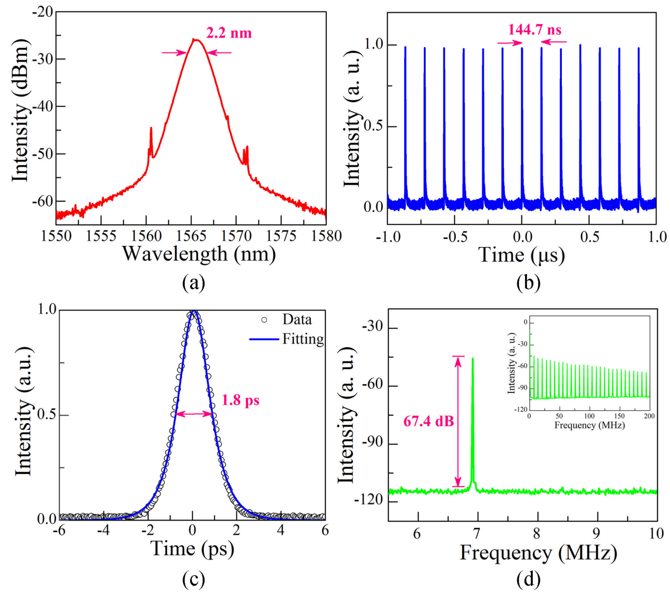

Fig. 4. (a) The spectrum, (b) the pulse train, (c) the autocorrelation trace, (d) RF spectrum (insert is the wide-band RF spectrum) of the soliton pulse of Er-doped fiber laser mode-locked by G-Bi_2Te_3-SA.

the pump power reach up to a mode-locking threshold of 30 mW. Fig. 4 shows the pulse characteristics at the pump power of 80 mW. The central wavelength of the mode-locking pulse is 1565.6 nm with a 3-dB bandwidth of 2.2 nm. Very clear symmetric spectral sidebands were observed, indicating that the mode-locked pulse is soliton pulse as shown in Fig. 4(a). The oscilloscope trace of the soliton pulse train had been plotted in Fig. 4(b) with the pulse-to-pulse time separation of 144.7 ns, corresponding to the repetition rate of the mode-locked pulse of 6.91 MHz. The pulse duration is 1.8 ps with fitting by sech^2-pulse profile, shown in Fig. 4(c). The real pulse duration is 1.17 ps by multiplying a factor of 0.65. The time-bandwidth product is 0.315. Fig. 4(d) shows the radio frequency (RF) spectrum of obtained soliton pulse. The fundamental repetition rate is 6.9 MHz corresponding to the cavity length of 28.99 m, the signal-to-noise ratio of the fundamental repetition rate is 67.4 dB. No addition frequency peak had been seen in the insert of the Fig. 4(d) with a wide-band RF spectrum up to 200 MHz.

For the broadband saturable absorption property of the G-Bi_2Te_3-SA, the mode-locked pulse is also obtained in 1 μm ring-cavity Yb-doped fiber laser by incorporating this SA inside the laser cavity. Fig. 5 shows the pulse characteristics of the Yb-doped fiber laser at a pump power of 115 mW. The central wavelength of the mode-locking pulse is 1049.1 nm with 3-dB bandwidth of 4.3 nm. The steep spectral edges have been observed, which is a typical shape of dissipative solitons, as shown in Fig. 5(a). The oscilloscope trace of the dissipative soliton pulse train has been plotted in Fig. 5(b) with a pulse-to-pulse interval of 265.6 ns, corresponding to a repetition rate of 3.7 MHz. The pulse duration is 144.3 ps. The RF spectrum of pulse is shown in Fig. 5(d). The fundamental repetition rate of the mode-locked pulse locates at 3.69 MHz with the signal-to-noise ratio of 65 dB, indicating that the cavity length is 54.20 m. No addition frequency peak had been seen with the wideband RF spectrum up to 100 MHz, shown in the insert of the Fig. 5(d).

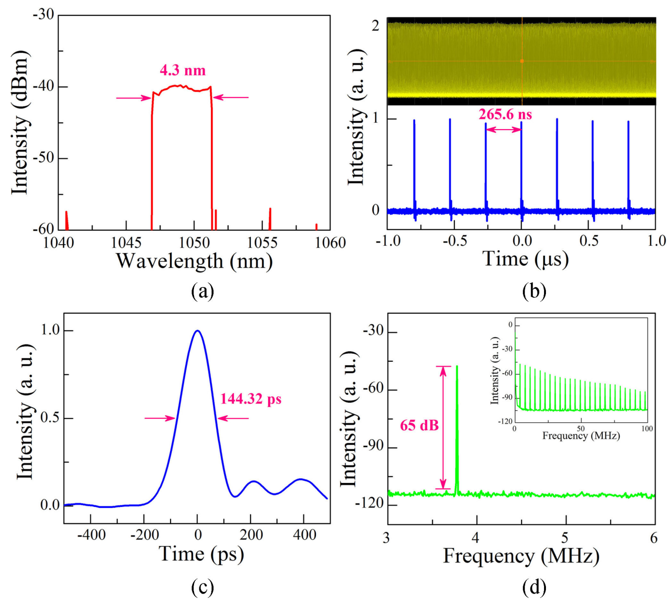

Fig. 5. (a) The spectrum, (b) the pulse train (insert is the large time scale), (c) the autocorrelation trace and (d) the RF spectrum (insert is the wideband RF spectrum) of the dissipative soliton of Yb-doped fiber laser mode-locked by G-Bi_2Te_3-SA.

III. Discussion

The graphene-Bi_2Te_3 heterostructure, made up of graphene and Bi_2Te_3 through Van der Waals interactions, keeps the linear energy-momentum "relativistic" dispersion of the electrons [52], which is responsible for broadband photonics operation. This paper firstly demonstrates the operation on both Er-doped and Yb-doped fiber laser mode-locked by the same G-Bi_2Te_3-SA with the bandwidth over about 500 nm, which may pave the way of fabricating broadband SA based on Dirac materials for mode-locking techniques.

It is worth commenting on the broadband performance of graphene SA [25], TI SA [45] and MoS_2 SA [44]. Due to low light absorption of graphene, the graphene-Bi_2Te_3 heterostructure SA have better nonlinear response than pure graphene SA, such as, the larger modulation depth and high damage threshold [48]. While the pure Bi_2Te_3 with heavily occupied by defects in the process of fabrication [53] exhibits its transport properties are dominated by the bulk state instead of the desired surface state [49]. The surface states of Bi_2Te_3 in graphene-Bi_2Te_3 heterostructure remain intact and are apparently well protected from environmental contamination with the help of graphene [51], indicated that broadband mode-locking operation can be more readily obtained by the G-Bi_2Te_3-SA than pure Bi_2Te_3 SA. Furthermore, the broadband MoS_2 SA with operation bandwidth dependent on the layer numbers of the MoS_2 film [31] needs more complex fabrications comparing to G-Bi_2Te_3-SA. Therefore, the G-Bi_2Te_3-SA remains very appealing for future broadband ultra-fast photonics.

IV. Conclusion

Herein, we have demonstrated a broadband SA of graphene-Bi_2Te_3 heterostructure for the passive mode-locking operation at both the Yb-doped and Er-doped fiber laser. The graphene-Bi_2Te_3 heterostructure is fabricated by two-step CVD processes. In comparison to pure graphene and pure Bi_2Te_3, graphene-Bi_2Te_3 heterostructure shows enhanced broadband absorption from 900 to 2000 nm. Based on the balanced twin-detector measurement system, the saturating intensity of 4.95 MW/cm^2 and modulation depth of 18.98% were measured at the communication band; while at 1067 nm, its saturating intensity and modulation depth were measured to be 2.61 MW/cm^2 and 23.11%, respectively. The ultrashort pulse with a duration of 1.1 ps and repetition rate of 6.9 MHz has been generated in Er-doped fiber at 1565.6 nm. Based on the Yb-doped fiber laser, the 144.3 ps pulses with repetition rate of 3.7 MHz at central wavlength of 1049.1 nm have been obtained. Our experimental findings provide another effective broadband optical material that may combine the advantages of graphene and topological insulator, which might finds potential applications beyond ultra-fast laser photonics.

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