Graphene-based Saturable Absorber for Pulsed Fiber Laser Generation

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Abstract: Recently, graphene has been considered as a great candidate to be applied as the saturable absorber (SA) with its brilliant optical characteristics such as ultrafast recovery time and ultra-wideband absorption due to its zero bandgap energy and linear dispersion of Dirac electrons. This paper focuses on reviewing the generation of short pulses from passive mode-locked fiber lasers that employ graphene-based saturable absorber (GBSA). Various parameters that make it excellent for generation ultra-short pulses including modulation depth, nonlinearity, saturation intensity, self-amplitude modulation, its crystal lattice structure, band gap energy distribution are discuss in details. Furthermore, comparison between single layers and multilayer GBSA is made to explain the effect of layers number on the behaviour of SA in ring cavity fiber lasers.

1. Introduction
Because the carbon element is comparatively plentiful and global in words in contrast to other type of elements and composites, different SA fabricated from carbon nanomaterials have been easily sophisticated with a convenient production and cost-effective issue. Such carbon nanomaterials have very attractive feature such as high thermal conductivity and excellent thermodynamic stability to act as optical element operate under high-optical-power with low damage, that’s particularly very suitable for using with relatively high-power passively mode locked fiber lasers.

Graphene is one of important carbon nanomaterial consider as an SP2 carbon atoms arranged in a layer as two dimensional honeycomb hexagonal lattice in which the charge carrier have massless Dirac fermion behavior and moved at ultrafast speed [1]. Graphene have many excellent properties which make it play important role in signal emitting, transmitting, detecting, modulating, in one material, such as high thermal conductivity, high optical damage threshold and high third-order optical nonlinearities[1, 2], ultra-fast carrier dynamic, and broadband optical absorption(2.3% per layer)[3].

In recent year, several investigations have reported about the fabrication of GBSA as passively mode locker element, such that, in 2010, Qiaoliang Bao et al, suggested monolayer graphene SA for mode locked fiber laser, the SA fabricated as Large area monolayer and multilayer graphene films that’s were grown by chemical vapor deposition (CVD) [3]. Also, in 2016, Zhang H. et al , proposed graphene mode locked fiber laser on which graphene SA act as reflection type saturable absorber for Erbium doped fiber laser (EDFL)[4]. In order to handle graphene sheet easily, solution-fabricated graphene can be gathered with a suitable type of polymers to produce a robust, transparent membrane [5]. This membrane combines the optical absorption of organics dye with polymer mechanical strength that’s reinforced composite with such dyes optical absorption. For example, in 2010, Zhipei Sun et al. made-up SA from graphene and polyvinyl alcohol (PVA) solution to generate mode locked pulse of 1
ps duration in C-band optical range [3]. In the same year, Qiaoliang Bao et al suggested the fabrication of a hybrid membrane of transparent graphene organic that could be handled with a tweezer [6].

This paper focuses on overview of the GBSA development for generating ultra-short pulses from passively mode-locked fiber laser (PMLFL) and reviewing the performance of single layer (SL) - GBSA and multi-layer (ML) -GBSA. The suitable operating parameters required for self-starting and stabilizing the process of mode locked within the ring fiber laser are listed for comparison and discussion. At first, various parameters that make GBSA excellent for generation ultra-short pulses including modulation depth (MD), nonlinearity, saturation intensity, self-amplitude modulation, its crystal lattice structure, band gap energy distribution are discuss in details. The basic characteristics of the SL-GBSA and ML-GBSA are discussed as well. In comparison, the SL-GBSA possess the best optical nonlinearity and acts as more effective SA, whereas the ML-GBSA thickness lead to increase non-saturable absorption and reduce modulation depth value, causing degradation in the performance of the SA in mode-locked operation. Finally, the use of SL-GBSA and ML-GBSA as a mode locker in PMLFLs are presented.

2. Graphene base saturable absorber GBSA

The graphene properties could be controlled by changing its chemical potential value ($\mu$), or the position of Fermi level and this can be made by either chemical doping or electrical gating because of its unique electronics structure that’s perform by conical-shaped conduction and valence bands coincide at the Dirac point. Its optical conductance is frequency independent in a broad band range of photon energies. The dynamic optical response of graphene could be estimate by using of Kubo formula [6] as follows:

$$\sigma = \sigma_{\text{intra}} + \sigma'_{\text{inter}} + \sigma^*_{\text{inter}}$$

Both interband conductivities $\sigma'_{\text{inter}} + \sigma^*_{\text{inter}}$ and intraband conductivities $\sigma_{\text{intra}}$ are function of chemical potential of graphene. Because of the existence of zero band gap energy in graphene, the contribution of both intraband and interband are competed and the transition within the band occurred only above the threshold value at which $|\mu| = \hbar \omega / 2$ [7]. The Graphene optical nonlinearities result from the interactions of incident optical field with phonons and electrons leading to a net dipole moment (polarization)[8], which could be described by a Taylor expansion [9] as:

$$P = \varepsilon_0 \left( \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 \right) + \cdots,$$  

where $\chi^{(1)}$ is the first order susceptibilities and $\chi^{(2)}$ and $\chi^{(3)}$ are the second- and third-order nonlinear susceptibilities, which is related to the nonlinear phenomena such as two-photon absorption, saturable absorption, self-focusing, and self-phase modulation, etc. Different physical process is affected by the nonlinear interband excitation, which are depended on several conditions of pumping process. For example, several quantum transitions happen under ultra-short optical pulses pumping, leading to nonlinear type of relaxation and recombination energy. Therefore, graphene is consider as promising material for many applications [8].

Third order susceptibility is the main reason beyond the most of graphene optical nonlinearities. Such as broadband saturable absorption on which optical losses of the light beam propagated through graphene could be decreasing at high intensity of incident optical signal [8]. Consequently; graphene shows great potential in ultra-short pulse generation and appearing as a very good saturable absorber for wide-range of passively mode locked operation. In which optical excitation of the interband transition generates a non-equilibrium population of carrier in both valance and conduction band in which optical absorption process will be saturated when a steady state happened between excited electrons and the relaxing electrons resulting from recombination process [10].
Because of the saturation intensity of GBSA in the visible band of optical spectrum approaching to its damage threshold the ultra-short pulse generation process in graphene faces many challenges, so, many author's and researcher groups suggested the using of GBSA in generation of ultra-short pulses in mode locked lasers in the telecommunication optical band[6, 11]. Basically the absorption coefficient of GBSA as function of the incident light intensity is given approximately by the following simple form[3, 40] as:

\[
\alpha = \alpha_0 \left(1 + \frac{I}{I_{sat}} \right)^{-1} + \alpha_{ns}
\]  

(3)

where \( \alpha_0 \) is the linear absorption coefficient, \( I_{sat} \) is the saturation intensity, and \( \alpha_{ns} \) is the non-saturated loss, and also, the nonlinear transmission of a GBSA can be describe by the following simple formula[6, 12]:

\[
T(I) = 1 - \Delta T \exp \left( - \frac{I}{I_{sat}} \right) - \alpha_{ns}
\]  

(4)

Where \( \Delta T \) is the modulation depth, \( I \) is the intensity of the input pulse, \( I_{sat} \) is the saturation intensity. One can note that the increasing of input intensity result increases of the SA transmission, which indicates the possibility of amplitude modulation of such device. Hence, the performance of GBSA could be determine by several parameters such as Modulation depth, Non-saturated optical losses, saturation intensity [6]. Figure1 shows an example of the SA transmission curve in which the modulation depth represents the difference between the non-saturated losses and the background absorption loss (\( \alpha_0 \))[3][39].

The GBSA could be fabricated as a thin film either as single layer or multi-layer or as polymer composite for passively mode locked fiber laser, using either physical or chemical methods on which number of layer could be known by different technique such as transmission electron microscopy (TEM), atomic force microscopy (AFM), and Raman spectroscopy (RS).

2.1 Single layer graphene based saturable absorber (SL-GBSA)

SLG have single layer of graphene nanoparticle with a honeycomb lattice structure that is formalized by SP2 hybridized carbon atoms in a planar configuration as shown in Figure 2 which possess a constant existence of electron-hole resonance carrier pair that’s have ultra-high drift velocity making them act as massless Dirac particles [13]. So that, it's optical inter-band transition is wave length independent and subtends only to the optical conductance of photon energies [14]. The SL-GBSA intrinsic properties make it the most successful SA for mode-locked fiber lasers compared to multilayer graphene. Excited carriers of SL-GBSA are expected to have ultrafast decay time because
of its zero band gap Dirac point [14]. However, the optical and electrical characteristics of thin graphene layer are highly sensitive to the existence of defects, sample quality and surface states [14].

The absorption characteristics of monolayer graphene, defective graphene and multilayer graphene have been examined [3] to show the effect of thickness of layer and defects existence on mode-locking performance in terms of saturation intensity, threshold intensity, modulation depth, recovery time and ability of pulse shaping. Accordingly, the monolayer graphene has showed a superior performance to those of defective or thicker graphene layers.

The SL-GBSA can be saturated at a relatively low excited level as compared to double and multi-layer GBSA reported with modulation depth around 67% and ultrafast relaxation time. Thus, SL-GBSA has been utilized for generating ultra-short pulses in mode-locking fiber laser [10]. Moreover, SL-GBSA could be used as reflection or transmission mode locker in different types of fiber laser systems due to its wide band of absorption [5]. For example, in 2010, reflection type of SL-GBSA for mode locked generation in Diode-pumped Er: Yb: glass laser with Pulse width of 260 fs, at 1550 nm has been reported [15]. Further, in 2012, Hyung Baek et. al. reported on high quality large-area transmission type SL-GBSA in Ti:Sapphire as passively mode-locked laser with ultrafast recovery times and excellent nonlinear absorption in 800 nm and pulse width of 63-fs [16]. In 2015, 91 fs pulses generated by Cr:YAG mode-locking laser at 1516 nm using a transmission type of SL-GBSA [17]. A high-quality transmission type of SL-GBSA for diode-pumped passively mode locked Tm:YAP laser operated at the center wavelength of 1988.5 nm with pulse width of 20 ns has been proposed [18].

Various methods for fabricating SL-GBSA have been described and realized depending on the required quality and applications. Recently, SL-GBSA has been fabricated either by graphene-polymer composites [5], or graphene sheets exfoliated from graphite in the liquid phase [19], mechanical exfoliation, and the CVD and transfer method [5]. In comparison to other methods, the CVD and transfer method could produce excellent-quality and GBSA with large-area under the required number of layers, which was considered as prefer one for the passive mode-locking pulse generation [20]. CVD could be satisfied either by thermally generated method or by plasma-enhanced method (PECVD). The PECVD has several advantages such as the shorter times of processing, faster growth and the chances of a relatively lower synthesis temperature. However, the quality of GBSA fabricated by PECVD is highly affected by power of plasma [21]. The thickness of fabricated graphene film could be identify by using Raman spectroscopy combined with optical contrast spectroscopy [11]. In view of final sophistication on SL-GB mode locked FLs, either the ring or the linear cavity base FL designs have been reported [5].

2.2 Multilayer graphene based saturable absorber (ML-GBSA)
ML-GBSA is a SA, which is composed from stack layers of graphene. The layer stacks could change the significant optical properties of graphene such as absorption characteristics and nonlinear properties and also its band gap. Despite several studies of ML-GBSA for ultrafast mode locked fiber laser[22], finite information is exist regarding the enhancement of number of graphene layer for the generating pulse duration and dynamics range in mode locked fiber laser. Since ML-GBSA is one of fast SA type, its nonlinear transmission could be described by simplified formula, in which the transmission is function of number of stack layer (N) as shown in the following equation [3]:

\[
\text{Transmission} = \frac{1}{1 + N} 
\]
\[ T (N) = \left(1 + \frac{1.13 \pi \alpha N}{2}\right)^{-2} \]  

Hence, the transmission of multilayer graphene film could be nearly estimate by measuring the number of layers (N) and the graphene fine structure constant (\(\alpha\)). The formula of equation (5) is valid for ML-GBSA with properly define sequence stacked. But in the case of undefined stacking, another formula must be used to describe the transmittance as shown in the following equation [3]:

\[ T (N) = (1 - \pi \alpha)^N \]  

Therefore, the measurement of the thickness of the graphene sheet is very important to determine its optical characteristics. Although, Atomic Force Microscopy (AFM) measurements represent one of the direct to find the N value, such method is slow and may cause damage to the internal structure during the process and also in such measurement the data fitting is needed to extract the true thickness of the graphene film. Recently, Raman spectroscopy is the most attractive candidate for fast and nondestructive checkup of the number of layer of the ML-GBSA. For example, Raman signature of single layer graphene include most of the features that are exist within all carbon-based material such as graphite, carbon nanotube, graphene oxide and reduced graphene oxide. For example Raman signature of single-layer graphene contained two distinct feature that one cannot see in even two-layer the first one is the small upshift of about 5 cm\(^{-1}\) of the G-peaks and the second is the improvement happen in graphite shoulder peak to second order D peak as shown in Figure 4[3]. And as the Raman spectrometer wavelength is altered to 633nm, this signature seem slightly noisy without appreciable change in the wave number value [23]. Also, Figure 4 shows the difference happened in Raman spectrum between single and multilayer of graphene structure [3, 22].

**Figure 3.** Raman signature of graphene and graphite
Raman spectroscopic methods is a possibility nominated for giving fast examination graphene thickness [22]. ML-GBSA film could be fabricated by the same method that’s mansions previously with the SL-GBSA film, but the CVD and transfer method is the most suitable one for the use with FLs because the number of graphene layers fabricated by such method could be easily control [24]. Such technique have several advantage such as it is very flexible and free from alignment and produced fully fiberized cavity making the laser system more compact, stable, and inviolable to external disturbances[24].

Recently, the fabrication technique is based on the effect of the evanescent field interaction that can be used with, side-polished (D-shaped) fibers, and tapered fibers (microfibers). In which the interaction between the deposited material and the propagating evanescent field of the cladding material has been exploited [25-27]. The multilayer GSA functioning with the evanescent field effect in fiber lasers has been investigated to generate an output pulses at 1561.1 nm with repetition rate of 6.99 MHz [28].

Principally, three basic nonlinear parameters of SA, namely: modulation depth, saturation intensity, and nonsaturable loss could be measured by power-dependent transmission setup [14]. Generally, in the fiber lasers, the modulation depth (MD) of a GBSA must be increased to some level in order to self-starting stable mode locking and produced ultra-short pulses. This might be done by scaling the number of layer in graphene SA. Also, MD is a critical parameter that limits the performance of the fiber laser [29, 30]. The modulation depth is changed by increasing the graphene layer to increase the non-saturated losses, which is not required in fiber laser [30].

Several researches have employed GBSA in mode locked fiber laser [30, 31]. These researches have proved that SL-GBSA acts as more effective saturable absorber in contrast to the ML-GBSA due to its intrinsic properties. For example SL-GBSA absorption could be saturated at lower value of excitation energy compare to multilayer one, and the modulation depth of SL-GBSA is larger effect than that of the ML-GBSA, because of both scattering and non-saturated loss occurs in the last one. Additionally, SL-GBSA has a better the pulse-shaping ability, output energy and pulse stability due to its larger modulation depth, ultrafast relaxation time and lower loss value. Also, surface defect and thickness lead to increase non-saturable absorption and reduce modulation depth value, causing degradation performance of the SA in mode-locked operation. Table 1 listed several experimental multi-layer and single-layer GBSA parameters with its fabrication methods.
Table 1. Summary of different mode locked GBSA parameters

| N  | Fabrication method | Centre wavelength (nm) | MD % | $\alpha_{ns}$ % | Saturation fluency ($\mu J/cm^2$) | Pulse duration (fs) | output power (mW) | Ref. |
|----|--------------------|-----------------------|------|----------------|-------------------------------|--------------------|-------------------|------|
| 1  | CVD                | 800                   | 1.8% | <0.9%          | 66.5                          | 63                 | 480               | [32] |
| 1  | CVD                | 1040                  | 0.75 | 1.59%          | 50                             | 160                | 160               | [33] |
| 1  | CVD                | 1250                  | 0.54 | 1.61%          | 14.5                           | 100                | 230               | [18] |
| 1  | CVD                | 1500                  | 0.5  | 1.9%           | 14                             | 91                 | 100               | [17] |
| 7  | CVD                | 1543.6                | 3.98 | 18.4%          | -                              | 1147               | 1.61              | [24] |
| 11 | CVD                | 1550.9                | 3.5  | 29.5%          | -                              | 715                | 1.7               | [24] |
| 12 | CVD as multilayer  | 1561.5                | 3.8  | 74.8%          | 10                             | 406                | 1.74              | [34] |
| 14 | CVD                | 1556.2                | 3.28 | 35.14%         | -                              | 563                | 1.7               | [24] |
| 21 | CVD                | 1559.12               | 2.93 | 53.05%         | 53.25                          | 483                | 1.99              | [24] |
| 24 | CVD as graphene/PMMA | 1560.34               | 5.6  | 62.6%          | 4                              | 350                | 2.3               | [34] |
| 37 | CVD as multilayer  | 1559.34               | 7.5  | 50%            | 3                              | 345                | 2.1               | [34] |
| 48 | CVD as multilayer  | 1557.0                | 9.6  | 39%            | 2                              | 359                | 1.5               | [34] |

3. Application of GBSA in passively mode locked ring cavity fiber laser

Mode locking by passive technique can be generated by incorporating of the SA into the ring cavity of the FL [38], such passive mode-locking technique is particularly very suitable to low-repetitive ultrashort pulse-train of fiber, if the carrier relaxation time of the SA is very short and it's modulation depth is relatively high which is dominates for the formation of transient net gain window, and the SA possess low saturation intensity with good pulse shaping ability [5]. Nowadays, there are different Carbone based Nano-material saturable absorbers (CBNMSAs) including single-walled carbon nanotubes (SWCNT), double-wall Carbone nanotube(DWCNT) graphene [37], Nano-graphite, graphene oxide, charcoal, which are developed for the mode-locked fiber lasers. In comparison to other CBNMSAs, the GBSA possess the suitable saturation intensity value with the relatively larger modulation depth value and shorter pulse duration with very good pulse shaping ability within the C-band of optical spectrum that is really important in optical communication system. Due to the linear dispersion of the massless Dirac fermions for the graphene nano-materials, the GBSA possesses the large optical linear absorption; in addition, all of its unique and excellent optical properties shown in the previous sections realize the development on various graphene-based saturable absorbers including SL-GBSA and ML-GBSA. Consequently, several investigation have been reported about the most brilliant characteristics of GBSA, including high nonlinearity and ultra-fast transportation, which are open a wide range of optical applications, specifically, it operates as passively mode locked in ring cavity fiber laser for short pulse generation[5].

In most cases the experimental set up of the mode locked erbium doped fiber laser (EDFL) employing the GBSA is shown in Figure 5. In which, Erbium doped fiber operated as gain medium that’s pumped optically by using diode laser. And the GBSA prepared as a thin film and incorporated as mode locker inside the ring cavity [35]. The suitable fabrication method and the suitable concentration with thickness of GBSA thin film are represent the key parameters to the production of stable mode-
locking pulses in such laser[36]. And a suitable length of single mode fiber (SMF) integrated into the laser cavity to compensate its dispersion and an auto correlator must be used to measure the actual pulse width of the mode-locked EDFL [36].

Finally, mode-locked fiber lasers based GBSA have been successfully demonstrated and they have great attraction. ML-GBSA have been proposed for mode locking of 1000 nm broadband ultrashort pulse generation from various fiber lasers [31]. Production of 1.23 ps laser pulses have been achieved by inserting SL-GBSA inside mode-locked EDFL [11]. Generation of 670 fs pulses with large spectral width of 4.99 nm using ML-GBSA for passively mode locked EDFL have been explored [6]. Moreover, EDFL based GBSA have produced of 174 fs pulses [38].

4. Conclusions

In presenting of previous development advancement on the graphene, nanomaterials base SA s, no matter if such SA constructed as single or multilayer for ultra-short pulse generation FLs, both of them are relatively cost-effective and could be easily produced and such type of carbon base nano material can play very important role on the initiating SAM of FL and using as base optical non-linear passive element for ultra-short pulse mode locking generation. In appointed, the PMLFL by GBSA having an operational wave length tuned widely within the range (1000-2500) nm for implementing to both near infrared NIR and infrared IR FL. By choosing suitable number of layer of the GBSA, the passively Q-switched FL could be able to switch from Q-switch operation to mode locked operation with pulse width variations from sub-ten of μs to hundreds of fs to be available for different short pulse applications. Finally, from general comparison between single layer and multi-layer GBSA, the single layer one could be consider the gold standard to produce lower saturation energy with a larger effective modulation depth, excellent pulse shaping ability and super-fast relaxation time, such optical characteristics dominated pulse suppression and formation in mode locked FL are the key parameters for the SAM effect. So that, With technological advancement on nano-material and structural modification for enhancement short pulse generation abilities, the GBSAs shall continue to be the super bright spotlight of cheap and convenient passive component for the FLs in the near future.

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