Q-Switching and Mode-Locking Pulse Generation in Ytterbium-Doped Fiber Lasers Using Nanomaterial Saturable Absorbers

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2017
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THESIS SUBMITTED IN FULFILMENT OF THE REQUIREMENTS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

FACULTY OF ENGINEERING
UNIVERSITY OF MALAYA
KUALA LUMPUR

2017
UNIVERSITY OF MALAYA
ORIGINAL LITERARY WORK DECLARATION

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Name of Degree: Doctor of Philosophy

Q-SWITCHING AND MODE-LOCKING PULSE GENERATION IN YTTERBIUM-
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Field of Study: PHOTONICS

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ABSTRACT

This research work focuses on exploring various new nanomaterials for saturable absorber (SA) application in generating Q-switched and Mode-locked pulses operating at 1 µm region. These nanomaterials are Molybdenum disulfide (MoS$_2$), Black Phosphorus (BP), Topological Insulator (TI): Bismuth (III) Selenide (Bi$_2$Se$_3$), Bismuth (III) Telluride (Bi$_2$Te$_3$), and antimony telluride (Sb$_2$Te$_3$), and metal oxide: Nickel Oxide (NiO) nanoparticles and cobalt oxide (Co$_3$O$_4$) nanocubes. The fiber laser employs Ytterbium-doped fiber (YDF) as a gain medium. Firstly, molybdenum disulfide (MoS$_2$) was proposed. The Q-switched laser was obtained by using few layers MoS$_2$, which was mechanically exfoliated by using a scotch tape. The SA was sandwiched between two fiber ferrules to form a fiber compatible Q-switcher. By incorporating the SA inside the YDFL cavity, a stable pulse laser operating at 1070.2 nm wavelength was generated with the repetition rate was tunable from 3.817 to 25.25 kHz. A passively mode-locked YDFL was demonstrated using a few layered MoS$_2$ film which was obtained by a liquid phase exfoliation technique. The mode-locking pulses have a repetition rate of 18.8 MHz and pulse energy of 0.1 nJ. Secondly, mechanically exfoliated Black phosphorus (BP) was proposed for both Q-switching and mode-locking pulses generation. The Q-switched laser has a pump threshold of 55.1 mW, a pulse repetition rate that is tunable from 8.2 to 32.9 kHz, the narrowest pulse width of 10.8 µs and the highest pulse energy of 328 nJ. BP based mode-locked YDFL was obtained by improving the SA preparation. The laser operated at 1033.76 nm with a fixed repetition rate of 10 MHz. Passively Q-switched YDFLs was also successfully demonstrated using a few-layers Bi$_2$Se$_3$, Bi$_2$Te$_3$ and
antimony telluride (Sb$_2$Te$_3$) based SAs. For instance, a Sb$_2$Te$_3$ film based Q-switched YDFL produced pulse repetition rate, which was tunable from 24.4 to 55 kHz with the maximum pulse energy of 252.6 nJ at 82.3 mW pump power. The mode-locked YDFL operating at 24.2 MHz repetition and 18.8 ps pulse width were also realized with Sb$_2$Te$_3$ based SA. Finally, two transition metal oxide nanomaterials: Nickel Oxide (NiO) and cobalt oxide (Co$_3$O$_4$) were embedded into a polymer film, making it an SA device for both Q-switched and mode-locked YDFLs. Stable Q-switched and mode-locked YDFLs were realized with both materials. For instance, the mode-locked Co$_3$O$_4$ based YDFL was operated at 1035.8 nm wavelength with a fixed repetition rate of 20 MHz and picoseconds pulse width. In short, an efficient and low-cost Q-switched and mode-locked YDFLs operating in 1 µm region have been successfully achieved by utilizing various new nanomaterials as SA.

**Keywords:** Q-switching, Mode-locking, Ytterbium-doped fiber (YDF), Saturable Absorber (SA).
PENJANAAN DENYUTAN Q-SUIS DAN MOD-KUNCI DALAM LASER GENTIAN BERDOP YTTERBIUM DENGAN MENGGUNAKAN BAHAN PENYERAP TEPU NANO

ABSTRAK

Penyelidikan ini tertumpu kepada kerja-kerja penerokaan bahan-nano baharu sebagai penyerap tepu (SA) bagi penghasilan denyut suis-Q dan mod-kunci yang beroperasi pada kawasan 1 μm. Bahan-nano ini adalah molibdenum disulfdida (MoS₂), fosforus hitam (BP), topologi penebat (TI): Bismut (III) selenida (Bi₂Se₃), Bismut (III) Telluride (Bi₂Te₃), dan telluride antimoni (Sb₂Te₃), dan logam oksida: Nikel oksida (NiO) zarah-nano dan kobalt oksida (Co₃O₄) kiub-nano. Laser gentian menggunakan gentian terdop-Ytterbium (YDF) sebagai medium gandaan. Pada awalnya, molibdenum disulfdida (MoS₂) telah dicadangkan. Laser suis-Q diperolehi dengan menggunakan beberapa lapisan MoS₂ yang telah dikelupas secara mekanikal dengan menggunakan pita lekat. SA telah diapit di antara dua ferrules gentian untuk membentuk gentian Q-pengsuisan. Dengan menggabungkan SA kedalam rongga YDFL, operasi denyut laser yang stabil telah dihasilkan pada panjang gelombang 1070.2 nm dengan kadar ulangan boleh laras dari 3.817 sehingga 25.25 kHz. YDFL mod-kunci pasif telah didemostrasikan dengan menggunakan filem berlapis MoS₂ yang diperolehi dengan menggunakan teknik pengelupasan fasa cecair. Denyut mod-kunci mempunyai kadar pengulangan 18.8 MHz dan tenaga denyut sebanyak 0.1 nJ. Pada bahagian kedua, BP yang dikelupas secara mekanikal telah dicadangkan untuk penjanaan denyut Q-suis dan mod-kunci. Laser Q-suis mempunyai ambang pam 55.1 mW, kadar ulangan denyut boleh laras dari 8.2 sehingga 32.9 kHz, lebar denyut yang paling sempit 10.8 μs dan tenaga denyut tertinggi 328 nJ. YDFL mod-kunci berasaskan BP diperolehi dengan meningkatkan mutu penyediaan SA. Laser beroperasi pada 1033.76 nm dengan kadar ulangan tetap pada 10 MHz. Pasif YDFL Q-suis juga berjaya dihasilkan menggunakan beberapa lapisan Bi₂Se₃,
Bi$_2$Te$_3$ dan telluride antimon (Sb$_2$Te$_3$) berasaskan SA. Sebagai contoh, filem Sb$_2$Te$_3$ berasaskan YDFL Q-suis menghasilkan kadar pengulangan denyut boleh laras dari 24.4 sehingga 55 kHz dengan tenaga denyut maksimum 252.6 nJ pada kuasa pam 82.3 mW. Operasi YDFL mod-kunci pada kadar ulang 24.2 MHz dan 18.8 ps lebar denyut juga direalisasikan dengan SA berasaskan Sb$_2$Te$_3$. Akhir sekali, dua peralihan logam oksida bahan-nano: Nikel Oksida (NIO) dan kobalt oksida (Co$_3$O$_4$) telah terbenam ke dalam filem polimer, menjadikannya peranti SA untuk kedua-dua YDFL Q-suis dan mod-kunci. YDFL Q-suis dan mod-kunci yang stabil dapat dicapai dengan kedua-dua bahan tersebut. Sebagai contoh, YDFL mod-kunci berasaskan Co$_3$O$_4$ telah beroperasi pada panjang gelombang 1035.8 nm dengan kadar ulangan tetap 20 MHz dan lebar denyut pikosaat. Secara ringkasnya, YDFL Q-suis dan mod-kunci beroperasi di kawasan 1 μm yang cekap dan murah telah berjaya dicapai dengan menggunakan bahan-nano baharu sebagai SA.

**Kata kunci:** Q-suis, Mod-kunci, Gentian Terdop-Ytterbium (YDF), Penyerap Tepu (SA).
ACKNOWLEDGEMENTS

I would like to express my greatest gratitude to my supervisors Prof. Ir. Dr. Sulaiman Wadi Harun and Prof. Dr. Hamzah Bin Arof for their continuous guidance and patience towards my research development process. They have guided me strictly to make sure that I could produce my best effort. They have spent their valuable time to discuss all problems and issues I had faced.

In addition, I would like to greatly thank my family for their continuous support and help, especially my parents who always care for my study and try to be patient for my absence, thanks to them for understanding and giving me this chance to complete my PhD research.

Last but not least, I want to take this opportunity and thank all my friends who gave me their words of encouragement and motivated me to finish my PhD research.

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LIST OF SYMBOLS AND ABBREVIATIONS

For examples:

LD : Laser Diode
UV : Ultra-voilet
Yb : Ytterbium
Yb$^{3+}$ : Ytterbium ion
YDF : Ytterbium-doped fibre laser
YDFL : Ytterbium-Doped Fiber Laser
EDFL : Erbium-Doped Fiber Laser
SA : Saturable Absorber
CVD : Chemical Vapour Deposition
LPE : Liquid phase exfoliation
CW : Continuous Wave
DMF : $N$-dimethylformamide
DI : Deionized
YDF : Ytterbium-Doped Fiber
WDM : Wavelength division multiplexer
FWHM : Full-Width at Half Maximum
FWM : Four-wave mixing
OSA : Optical Spectrum Analyzer
OSC : Oscilloscope
OPM : Optical power meter
SNR : signal-to-noise ratio
PC : Polarization Controller
PVA : Polyvinyl Alcohol
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>PEO</td>
<td>Polyethylene Oxide</td>
</tr>
<tr>
<td>RE</td>
<td>Rare-Earth</td>
</tr>
<tr>
<td>SDS</td>
<td>Sulphur Dodecyl Sulfate</td>
</tr>
<tr>
<td>SMF</td>
<td>Single Mode Fiber</td>
</tr>
<tr>
<td>SRS</td>
<td>Stimulated Raman Scattering</td>
</tr>
<tr>
<td>NPR</td>
<td>Nonlinear Polarization Rotation</td>
</tr>
<tr>
<td>NA</td>
<td>Numerical aperture</td>
</tr>
<tr>
<td>GVD</td>
<td>Group Velocity Dispersion</td>
</tr>
<tr>
<td>MOCVD</td>
<td>Metal-organic chemical vapor deposition</td>
</tr>
<tr>
<td>RF</td>
<td>Radio Frequency</td>
</tr>
<tr>
<td>TBP</td>
<td>Time-Bandwidth Product</td>
</tr>
<tr>
<td>1D</td>
<td>One-Dimensional</td>
</tr>
<tr>
<td>2D</td>
<td>Two-Dimensional</td>
</tr>
<tr>
<td>SESAMs</td>
<td>Semiconductor Saturable Absorber Mirrors</td>
</tr>
<tr>
<td>CNTs</td>
<td>Carbon Nanotubes</td>
</tr>
<tr>
<td>TMDs</td>
<td>Transition Metal Dichalcogenides</td>
</tr>
<tr>
<td>MoS$_2$</td>
<td>Molybdenum disulfide</td>
</tr>
<tr>
<td>PB</td>
<td>Black Phosphorus</td>
</tr>
<tr>
<td>TIs</td>
<td>Topological Insulators</td>
</tr>
<tr>
<td>Bi$_2$Se$_3$</td>
<td>Bismuth (III) Selenide</td>
</tr>
<tr>
<td>Bi$_2$Te$_3$</td>
<td>Bismuth (III) Telluride</td>
</tr>
<tr>
<td>Sb$_2$Te$_3$</td>
<td>Antimony Telluride</td>
</tr>
<tr>
<td>Te</td>
<td>Tellurium</td>
</tr>
<tr>
<td>NiO</td>
<td>Nickle Oxide</td>
</tr>
<tr>
<td>Co$_3$O$_4$</td>
<td>Cobalt (II, III) oxide</td>
</tr>
<tr>
<td>TiO$_2$</td>
<td>Titanium dioxide</td>
</tr>
</tbody>
</table>
E_v : Energy level of valance band
E_c : Energy level of conduction band
T_BL : Time Bandwidth Product
Sech^2 : Hyperbolic-secant-squared
XRD : X-ray Powder Diffraction
TEM : Transmission electron microscopy
FESEM : Field Emission Scanning Electron Microscopy
GNS : Graphene nanosheets
SML : Self-mode locking
TDFL : Thulium doped fiber laser
CHAPTER 1: INTRODUCTION

1.1 Overview of Laser and Pulse Fiber Laser

Lasers have many applications in our daily life, such as being used in households’ appliances, hospitals’ tools and communication means. Fiber lasers are one type of lasers that have undergone a lot of development efforts in the last two decades. They have become the main focus of interest to many researches, due to their high reliability, improved beam quality, lower noise floor and higher power application; as compared to the solid state and semiconductor lasers. Laser is found in various industrial applications such as material processing, optical communications, spectroscopy and imaging (Siegman, 1986; H. Zhang et al., 2009). The laser operating regimes can be classified on the basis of temporal characteristic of optical output power. Concentrating the available energy in a single, short optical pulse, or in a periodic sequence of optical pulses; transient laser behavior can generate higher peak power than the one shown by the continuous wave (CW) laser. The most important regimes are continuous wave (CW), Q-switching, mode-locking, and Q-switched mode-locking.

Fiber lasers have a broad range of applications ranging from industry to optical communication. Various laser setup generates pulse with different and distinctive pulse characteristics. Subsequently, each laser setup can be tailored to accommodate for one or some other applications. For an instance, Q-switched laser with high peak intensity and high pulse energy is suitable for micromachining and drilling which benefits the medical, electronic and automotive industry (Nikumb et al., 2005). Q-switched lasers are also used in the medical fields especially in eye and dental surgery (Plamann et al., 2010; Serbin et al., 2002). Eye surgery is one of the applications of pulsed laser by which the system is known as the laser-assisted in situ keratomileusis (LASIK) (Kezirian et al., 2004). In LASIK, ultra-voilet (UV) laser is also used to photo-ablate the corneal tissue rather than mechanical cutting since the later will somehow damage the surface layer or cornea and
the surrounding cells. Moreover, Q-switched laser is invested heavily to mark information such as batch number, manufactured date and logo in the electronic semiconductor manufacturing industry (Noor et al., 1994). On the other hand, mode-locked lasers can be used to generate ultra-short pulses train. Ultra-short pulsed fiber lasers have attracted widespread interests because of their useful applications in many areas such as micromachining, communication, and optical systems (Bao et al., 2009; Norihiko Nishizawa, 2014).

Both Q-switched or mode-locked fiber lasers can be generated by either active or passive techniques. The active technique was usually realized by integrating an external controller device such as acoustic-optic modulator to actively modulate the light of the intracavity (Bello-Jiménez et al., 2010). On the opposite side, the passive technique utilizes saturable absorption of optical material to change the intra-cavity light and generates pulsed laser in the cavity (De Tan et al., 2010). Passive Q-switching laser operation depends on the gain medium as well as the saturable absorber (SA) which acts as a Q-switcher and it occurs when the photon flux starts to show a gain, fixed loss and saturable loss in the SA after many round-trips. The first SA was demonstrated in 1966 to generate pulses in Nd: glass laser (Stetser et al., 1966).

Passive mode-locking uses SA in a laser cavity to produce pulses of light of extremely short duration, on the order of nanoseconds to picoseconds. The basis of the technique is to induce a fixed-phase relationship between the longitudinal modes of the laser's resonant cavity. The repetition rate is defined as a free spectral range of cavity in MHz for few meter fiber laser cavity (H. Zhang et al., 2010). The main point for generation mode-locking fiber laser is to get higher repetition rate. The passive Q-switched and mode-locking method has become a commonly explored in recent years because of its simplicity as opposed to the active method (Grelu et al., 2012; A. P. Luo et al., 2011; H. Zhang et
To date, many passive techniques are proposed and demonstrated to generate Q-switched and mode-locked fiber lasers. Therefore, this study aimed to demonstrate Q-switched and mode-locked fiber lasers operating at 1 micron region using new types of SAs.

1.2 Recent Progress in Saturable Absorber Technology

Saturable absorption is a property of materials where the absorption of light decreases with increasing light intensity. Most materials show some saturable absorption, but often only at very high optical intensities (close to the optical damage). At sufficiently high incident light intensity, atoms in the ground state of a saturable absorber (SA) material become excited into an upper energy state at such a rate that there is insufficient time for them to decay back to the ground state before the ground state becomes depleted, and the absorption subsequently saturates. Figure 1.1 shows a simple description of the basic principle of the SA that is based on energy levels of the SA material for the valence band ($E_v$) and conduction band ($E_c$). Electrons in the valence band are excited up to the conduction band by the incident light. Most of photons are absorbed for low intensity and negligible number for high intensity due to the occupation of electronic density of states in the conduction bands. Thus, the saturable absorption appears depending on the transparency of the optical intensity.

Figure 1.1: Schematic diagram of the saturable absorber’s principle for (a) low incident light intensity (b) high incident light intensity
For an optical fiber system, the SA is inserted into the laser cavity. The light is passed through the SA in both high and low intensities. A high proportion of constituent photons in low-intensity light will be absorbed by the electrons at the valance band then excite of these electrons to the conduction band of the SA. In the other hand, the absorbance of the photons will decrease at high intensity. During each round trip, an intensity-dependent attenuation is created and appears in the low loss for high intensity and high loss for low intensity leading to not passing of an optical pulse through the SA (Kashiwagi et al., 2010). Therefore, the passed light through the saturable absorber leads to high-intensity contrast and starts to oscillate in the pulsed state. The key parameters for a saturable absorber are its wavelength range (where it absorbs), its dynamic response (how fast it recovers), and its saturation intensity and fluence (at what intensity or pulse energy it saturates). They are commonly used for passive Q-switching and mode-locking.

Up to date, there are various kind of SAs have been proposed such as semiconductor saturable absorber mirror (SESAM) (M. Zhang et al., 2009), graphene (Z. Sun et al., 2010), and carbon nanotube (CNT) (Della Valle et al., 2006). SESAMs were widely used since they have excessive flexibility, stability, and fast amplitude modulation. However, they have many drawbacks such as greater pricey, tough fabrication strategies, narrower wavelength operation range and contain a limited bandwidth of optical response (Ursula Keller et al., 1996). Thus, other SA materials have gained the attention of researchers due to their high overall performance, cheaper and broadband operation in the optical laser. In more recent years, CNTs and graphene had been commonly investigated in understanding all pulse fiber lasers due to their extensive absorption range and proper compatibility with optical fibers. However, CNTs and graphene have comparatively low modulation depth, which restricted their application for high power pulsed fiber lasers (Ahmed et al., 2014; Tolstik et al., 2014).
A new family of 2D nanomaterials, such as Black Phosphorus (BP) and molybdenum disulfide (MoS$_2$), is also drawing more interest for both Q-switching and mode-locking fiber laser applications. Topological insulators (TIs) have been shown to be capable of generating Q-switched and mode-locked lasers (Y. Chen et al., 2014; H. Liu, X.-W. Zheng, et al., 2014). Since TIs have a large modulation depth with an efficient saturable absorption property, they are suitable for making SAs. Very recently, a new kind of metal nanoparticles material such as gold, silver nanoparticles and titanium dioxide (TiO$_2$) were also investigated and demonstrated as a SA for Q-switched and mode-locked fiber laser (H Ahmad, Siti Aisyah Reduan, et al., 2016; Glubokov et al., 2014; T. Jiang et al., 2013).

In this thesis, four types of passive SAs; Black phosphorus, MoS$_2$, TIs and transition metal oxide are explored for generating Q-switching and mode-locking pulses in Ytterbium-doped fiber laser (YDFL) cavity.

1.3 Research Objectives

The aim of this work is to fabricate and demonstrate an efficient and low-cost Q-switched and mode-locked YDFLs operating in 1 µm region by utilizing new nanomaterials as SA. The following objectives have been outlined to achieve the aim;

1. To fabricate and characterize several types of nanomaterials based SAs, Molybdenum disulfide (MoS$_2$), Black Phosphorus (BP), Topological Insulators (TIs), Nickle Oxide (NiO) nanoparticles, and Cobalt oxide (Co$_3$O$_4$) nanocubes.

2. To demonstrate the capability of the fabricated materials saturable absorber (SA) to generate high pulse energy of Q-switched Ytterbium-Doped Fiber Laser.

3. To investigate and demonstrate the generation of the mode-locking pulses in YDFL cavity using the fabricated nanomaterials as SA.
1.4 Overview of the Thesis

This thesis is organized into seven main chapters, which report the comprehensive study of Q-switched and mode-locked pulse laser generations using several types’ nanomaterial as SAs. The current Chapter gives a brief introduction on the recent development of pulsed fiber lasers and SAs as well as highlighting the motivation of this work. Besides, the objectives of this study are also described. Chapter 2 briefly describes the overview of fiber laser technologies especially for operation at 1 μm region and provides a detailed of several new materials used to generate pulses fiber lasers. The description on Q-switched and mode-locked methods using several types of nanomaterial as passive SAs are also discussed in this chapter.

Chapter 3 demonstrates both Q-switched and mode-locked fiber lasers using ytterbium-doped fiber (YDF) as a gain medium and MoS$_2$ as SA. Firstly, a Q-switched YDFL is demonstrated using a few layers of MoS$_2$, which are mechanically exfoliated from a natural MoS$_2$ crystal using a scotch tape. The SA is sandwiched between two fiber ferrules to form a fiber compatible Q-switcher. Secondly, a mode-locked YDFL is also realized using another MoS$_2$ based SA, which was realized by embedding the MoS$_2$ into a polymer film.

In Chapter 4, both Q-switching and mode-locking operations in 1 μm region are demonstrated using Black Phosphorus (BP) as SA. The first sub-chapter describes a Q-switched YDFL using a newly developed multi-layer BP SA. The second sub-chapter describes the performance of mode-locking by incorporating the BP SA into the YDFL cavity as SA. Chapter 5 is divided into two main sub-chapters. In the first sub-chapter, the comparison of Q-switching performance in YDFL cavity by using two types of topological insulator TI: Bismuth (III) Selenide (Bi$_2$Se$_3$) and Bismuth (III) Telluride.
(Bi$_2$Te$_3$) as SA, is described. In the second sub-chapter, Antimony telluride (Sb$_2$Te$_3$) SA was used for generating Q-switching and mode-locking pulses train in YDFL cavity.

Two new types of transition metal oxide nanomaterials; laser; Nickel Oxide (NiO) and cobalt oxide (Co$_3$O$_4$) are explored as SA for Q-switching and mode-locking operations in 1 μm region. These materials are embedded into a polymer film and sandwiched between two fiber ferrules, making it an SA device. Chapter 7 provides the summary, analysis and review of all the results achieved in this study.
CHAPTER 2: LITERATURE REVIEW

2.1 Introduction

This chapter begins with a brief overview of the fiber laser technology for 1-micron region, which describes the ytterbium doped fiber (YDF) as the gain medium. Passive laser pulse formation is also presented in this chapter, which divided into two techniques (Q-switching and mode-locking). Nonlinear effects in optical fiber and saturable absorption are also described in this chapter. The enhanced focus is given to saturable absorber (SA) devices, where a few new materials such as the transition metal dichalcogenides (TMDs), Black phosphorous (BP), Topological insulator (TIs) and transition metal oxide are briefly described. The fabrication techniques of these SA materials are also covered in this chapter.

2.2 Fiber Laser Technology for 1-micron Region

Fiber lasers have gained more attention by many researchers as a promising laser configuration after it was firstly discovered in 1961 (Koester et al., 1964; Snitzer, 1961). Lasers were played an attractive role in the development of photonic technologies since the first laser demonstration by Maiman (Maiman, 1960). Lasers use the quantum impact of excited emission to produce light and they are constructed based on the following elements; an active medium as a gain provider, a pumping source to produce the energy and an optical cavity to reinforce and control the optical field (Agrawal et al., 1986). Ytterbium-doped fiber laser (YDFL) is the most efficient fiber lasers, which has many applications in various areas.

Ytterbium (Yb) is a chemical element that belongs to the group of rare earth metals. It is used as a laser active dopant in the form of the trivalent ion Yb$^{3+}$ to generate laser in 1 micron region. Figure 2.1 shows a sub-level Strark splitting in energy diagram of Yb$^{3+}$ ions and the sub-level splitting is depended on the position and concentration of Yb$^{3+}$
glass (Barua et al., 2008; Y. Qiao et al., 2008). In other words, Yb$^{3+}$ has a very simple electronic level structure, with only one excited state manifold ($^2F_{5/2}$) within the reach from the ground-state manifold ($^2F_{7/2}$) as seen in the same figure.

Figure 2.2 shows the ytterbium-doped fiber (YDF) cross-section of emission and absorption (Y. Qiao et al., 2008). Ytterbium displays a broadband absorption spectrum becoming multi-wavelength pumping and then turning into a high power operation. This wide absorption spectrum additionally permits the utilization of unstabilized and low-cost pumps, providing the design and lower cost in overall as well as a stable high power fiber lasers in long-term. The simple electronic structure excludes excited-state absorption and also a variety of detrimental quenching processes. The upper state lifetimes are typically in the order of 10 to 15 fs, which is beneficial for Q-switching and mode-locking pulses. The Yb$^{3+}$ ion possesses a number of emission transitions within the 950 – 1100 nm wavelength range. Furthermore, the homogeneous and inhomogeneous broadening of these transitions within a glass host, leads to a wide and continuous emission spectrum in the 1 micron band, as shown in Figure 2.2. The lifetimes of emission and absorption spectrum depend on the host materials (Weber et al., 1983).

The broadened absorption and emission transitions that make up the above spectrum occur between sublevels of the ground and the first excited states of Yb. Note that Yb degeneracy was lifted due to the Stark effect. The electronic configuration of Yb$^{3+}$ is [Xe] 4f$^{13}$, which results in the following values for the orbital, spin and total angular momentum quantum numbers: $L = 3$, $S = \frac{1}{2}$ and $J = \frac{5}{2}, \frac{7}{2}$.

Applying Hund’s rules produces the term symbols $^2F_{7/2}$ for the ground state as well as for the excited state (Griffiths, 2016). In the absence of an external perturbation, each of these states further consists of $2J+1$ degenerate (equal energy) angular momentum states, corresponding to the allowed values of $m_J$. 
Figure 2.1: Sub-level Strark splitting in energy diagram of Yb$^{3+}$ ions

Figure 2.2: Typical emission and absorption spectra of Ytterbium doped fiber (YDF)
Applying Hund’s rules produces the term symbols $^2F_{7/2}$ for the ground state as well as for the excited state (Griffiths, 2016). In the absence of an external perturbation, each of these states further consists of $2J+1$ degenerate (equal energy) angular momentum states, corresponding to the allowed values of $m$. The degeneracy of these states may be lifted through the application of an external electric field, which turns each set of the degenerate states into a manifold of Stark levels. The external field polarizes the atom and thereafter interacts with the resulting dipole moment as per to the interaction potential ($EV$). Since the interaction is dependent on only the magnitude of $m$, it consists of $J+\frac{1}{2}$ Stark levels. As for the case of Yb$^{3+}$, this implies that the $^2F_{7/2}$ ground state manifold is split into four Stark levels and the $^2F_{5/2}$ excited state is split into three Stark levels. It was shown that the absorption transitions between each of these manifolds occur from the lowest energy ground state Stark level, terminating each of the three excited state Stark levels (See Figure 2.3) (S. Dai et al., 2002; C. Jiang et al., 2000). The fluorescence transitions begin at the lowest energy $^2F_{5/2}$ Stark level and terminate on each of the $^2F_{7/2}$ Stark levels. These three absorption lines and four emission lines are then homogeneously and inhomogenously broadened due to their presence in an amorphous glass host, which forms continuous absorption and emission spectra, as shown in Figure 2.2.

Figure 2.3: Stark levels and absorption and fluorescence transitions for Yb$^{3+}$. The transitions are labeled on the terminating energy level with the absorbed or emitted photon wavelength. Values for the absorbed or emitted wavelengths obtained from (S. Dai et al., 2002)
The cross-sections of the emission and absorption are also known as the range of efficiency in fiber laser system. At the side of the fiber parameters, the emission and absorption focus on the power extraction efficiency and signal saturation energy. The chosen pump wavelength depends on the heat dissipation during the quantum defect, absorption and total fiber length. Figure 2.4 shows the schematic efficiency of YDFLs. Besides the quantum defect, there are impossible to avoid the loss contribution from excess pump, signal losses, and non-optimized cavity. By selecting convenient core and cladding materials and a suitable design, these losses can be minimized (Zervas, 2014).

![Output efficiency](image.png)

**Output efficiency**

- 60-80%
- Non-optimized cavity: 0.5-1%
- Excess signal loss: 5-10%
- Excess pump loss: 5-10%
- Quantum defect: 8-12%

**Figure 2.4: The schematic efficiency of YDFLs (Zervas, 2014)**

The field applied to the Yb$^{3+}$ ions is highly dependent on the glass host composition as well as any co-dopants and their concentrations. As a result, it is possible to control both, the transition wavelengths as well as the transition strengths, to a large degree by altering these properties (Barua et al., 2008). A large number of ytterbium-doped glasses are now available commercially in both bulk and fiber form. In this thesis, a commercial YDF, which was drawn from ytterbium-doped phosphosilicate glass, is used as a gain fiber. The operating regimes of the laser can be classified on the basis of temporal characteristics of output emission.
2.3 Pulse Formation in Fiber Laser Cavity

Short and ultrafast pulsed fiber lasers have broadly drawn in enthusiasm in recent years because of their application advantages, for example, in areas of communication, medicine and micromachining and additionally inclination being compact, simple to setup and cost-effective (Grelu et al., 2012; Sotor, Sobon, Macherzynski, et al., 2014). These lasers could be Q-switched or mode-locked fiber lasers, which can be generated by either active or passive techniques. The active technique was usually realized by integrating an external controller device such as acoustic-optic modulator to actively modulate the light of the intracavity (Bello-Jiménez et al., 2010). On the opposite side, the passive technique utilizes saturable absorption of optical material to change the intra-cavity light and generates pulsed laser in the cavity (De Tan et al., 2010). This section explains about the working principles of Q-switching and mode-locking operation and reviews about the previous works on both lasers.

2.3.1 Q-Switching Technique

Q-switching is a technique to achieve high energy laser pulses by inserting an intra-activity loss to modify the quality factor $Q$ of the resonator. The $Q$-factor expression equation is shown as follows; (Svelto et al., 1998)

$$ Q = \frac{2\pi E_s}{E_l} $$

(2.1)

where $2\pi$ as a product, $E_s$ is the ratio of stored energy in the resonator, and $E_l$ is the loss energy per resonator cycle. Passive Q-switching laser operation depends on the gain medium as well as the saturable absorber (SA) which acts as a Q-switcher and it occurs when the photon fluxes start to show gain, fixed loss and saturable loss in the SA after many round-trips.
The Q-switching pulse output performances are analyzed by parameters, such as pulse width, repetition rate, peak power, and pulse energy. The range of pulse repetition rate is commonly in kHz and pulse width in µs. Comparison to mode-locking technique, Q-switching has comparatively for much longer pulse width, far lower in repetition rate that matching to the time among two sequential pulses for restoring the emitting energy and depend on the electron’s lifetime in the excitation state inside the gain medium. The advantages of Q-switching technique is easier to achieve the pulses with no reaching the equilibrium between nonlinearity and dispersion of the medium, which is necessary for mode-locking technique (Popa et al., 2011).

A passively Q-switched laser includes a SA into the cavity and thus no external modulator is needed and the cavity loss is modulated by the SA to obtain a Q-switching pulses train. The inclusion of SA modulates the Q factor to periodically emit light as a pulse train with a kHz repetition rate range and pulse width ranging from ns to µs. SA can also be exhibited artificially by polarization effect, such as nonlinear polarization rotation (NPR) (Z.-C. Luo et al., 2012). So far, many passive SAs are also reported for Q-switching such as single and multiple walled carbon nanotubes (Ahmed et al., 2015), graphene (Z. Luo et al., 2010), topological insulator (Z. Yu et al., 2014) and Molybdenum Disulfide (MoS$_2$) (Z. Luo et al., 2014).

The SA transmission or reflection depends on the light intensity, where low light intensity will be absorbed by the material and high light intensity will be released depending on the material recovery time. For instance, Q-switched YDFL was reported using topological insulator Bi$_2$Se$_3$ SA to generate pulses with a repetition rate ranging from 8.3 to 29.1 kHz (Z. Luo et al., 2013). A few-layer molybdenum di-selenide (MoSe$_2$) SA was also used for Q-switching generation in 1 μm region (Z. Luo et al., 2013; Woodward et al., 2015). More recently, black phosphorus (BP) based SA was also used
to realize a tunable Q-switched YDFL with a pulse energy of 7.1 nJ (Harith Ahmad et al., 2016).

<table>
<thead>
<tr>
<th>Pulse laser</th>
<th>Materials</th>
<th>Max pulse energy (nJ)</th>
<th>Min pulse duration (µs)</th>
<th>Repetition rate (kHz)</th>
<th>λ (nm)</th>
<th>ΔT (%)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>YDF</td>
<td>CNTs</td>
<td>143.5</td>
<td>12.18</td>
<td>7.9–24.27</td>
<td>1060.2</td>
<td></td>
<td>(Kasim et al., 2014)</td>
</tr>
<tr>
<td>YDF</td>
<td>CNTs</td>
<td>18.4</td>
<td>1</td>
<td>30-50</td>
<td>1061</td>
<td></td>
<td>(Li et al., 2013)</td>
</tr>
<tr>
<td>YDF</td>
<td>Graphene</td>
<td>46</td>
<td>~70 ns</td>
<td>140–257</td>
<td>1064.2</td>
<td>20</td>
<td>(J. Liu et al., 2011)</td>
</tr>
<tr>
<td>YDF</td>
<td>Graphene</td>
<td>141.8</td>
<td>1.3</td>
<td>28.9-110</td>
<td>1027</td>
<td>8</td>
<td>(L. Zhang et al., 2012)</td>
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<tr>
<td>YDF</td>
<td>MoS₂</td>
<td>6.9</td>
<td>2.2</td>
<td>58-105</td>
<td>1068.2</td>
<td></td>
<td>(R. I. Woodward et al., 2014)</td>
</tr>
<tr>
<td>YDF</td>
<td>MoS₂</td>
<td>32.6</td>
<td>5.8</td>
<td>6.4-28.9</td>
<td>1066.5</td>
<td>1.6</td>
<td>(Z. Luo et al., 2014)</td>
</tr>
<tr>
<td>YDF</td>
<td>MoSe₂</td>
<td>116</td>
<td>2.85</td>
<td>60- 74.9</td>
<td>1060</td>
<td>4.7</td>
<td>(Woodward et al., 2015)</td>
</tr>
<tr>
<td>YDF</td>
<td>BP</td>
<td>2.09</td>
<td>1.16</td>
<td>52.52-58.73</td>
<td>1038.68 &amp; 1042.05</td>
<td>73</td>
<td>(Rashid et al., 2016)</td>
</tr>
<tr>
<td>YDF</td>
<td>BP</td>
<td>7.1</td>
<td>4</td>
<td>6.0–44.8</td>
<td>1056.6-1083.3</td>
<td>73</td>
<td>(Harith Ahmad et al., 2016)</td>
</tr>
<tr>
<td>YDF</td>
<td>Bi₂Se₃</td>
<td>17.9</td>
<td>1.95</td>
<td>8.3–29.1</td>
<td>1067</td>
<td>3.8</td>
<td>(Z. Luo et al., 2013)</td>
</tr>
<tr>
<td>YDF</td>
<td>Bi₂Se₃</td>
<td>6.2</td>
<td>2.1</td>
<td>14.9–62.5</td>
<td>1050.4</td>
<td>39.8</td>
<td>(Haris et al., 2017)</td>
</tr>
<tr>
<td>YDF</td>
<td>Bi₂Te₃</td>
<td>38.3</td>
<td>1</td>
<td>35-77</td>
<td>1056</td>
<td>2.5</td>
<td>(Junsu Lee, Koo, Chi, et al., 2014)</td>
</tr>
</tbody>
</table>
Table 2.1 compares several Q-switched YDFLs, which were obtained using 2D nanomaterials and other common materials. The maximum pulse energy of 141.8 nJ was successfully achieved using graphene based SA (L Zhang et al., 2012). This high energy pulse laser is suitable for applications in sensing, communication, and material processing. The wavelength of operation for the Q-switched YDFLs is 1027 nm (L Zhang et al., 2012) and 1083.3 nm (Harith Ahmad et al., 2016). The maximum repetition rate and pulse duration were 257 kHz (J. Liu et al., 2011) and 1 µs (Junsu Lee, Koo, Chi, et al., 2014; Li et al., 2013) respectively. Among those materials, the maximum modulation depth of 73% was obtained with the BP SA (Harith Ahmad et al., 2016; Rashid et al., 2016). The high modulation depth is advantageous to suppress the wave-breaking and produces high pulse energy.

2.3.2 Mode-locking Technique

Mode-locking is a technique to generate an ultrashort pulse laser. It can be realized using a passive technique based on SA. An ultrashort pulse can emerge when a SA modulates the loss once per cavity round-trip and all longitudinal modes have a fixed phase relationship. Thus, the mode-locking of the oscillating laser produces an ultrashort pulses train (ranging from ns to fs duration) at a defined repetition rate in MHz corresponding to the free spectra range of laser cavity or the number of obtained pulses per second (H. Zhang et al., 2010). The estimation of pulse repetition rate, $f$ in passive mode-locking technique is given by:

$$Repetition \ rate, f (ring \ cavity) = \frac{c}{nL}$$  \hspace{1cm} (2.2)

where $c$, $n$, and $L$ denote the speed of light ($3 \times 10^8 m/s$), refractive index of the medium (1.46 for silica fiber), and total cavity length, respectively. It is shown from the equation that the repetition rate is determined by the total cavity length for a passive
mode-locking, and therefore, the higher pulse repetition rate is obtained for a shorter cavity length.

The pulse width of the laser indicates the full width at half maximum (FWHM) of the power versus time and a very short pulse width can be realized by a mode-locked laser. The higher numbers of longitudinal modes that have a fixed phase relationship can translate to a shorter pulse width. The short pulse duration of the mode-locking mode is useful for many applications including fast optical data transmission, and time-resolving process. The relationship between pulse width and bandwidth of the optical fiber pulses is referred to a time-bandwidth product (TBP). As described by the principle of Heisenberg, the TBP of the pulse is impossible to drop below a limit of $T_{BL}$,

$$ T_{BL} \leq \Delta t \times \Delta v \quad (2.3) $$

where $\Delta t$ and $\Delta v$ denote the temporal width (in seconds) and the spectral width (in hertz) of the pulse, which measured at FWHM. The limit of TBP or $T_{BL}$ is depended on the pulse shape as described in Table 2.2. The bandwidth of the pulse is depended on the spectral bandwidth and operating wavelength of the output spectrum of the laser. It is given as;

$$ The \ bandwidth \ (BW) = \Delta \lambda \times \frac{c}{\lambda^2} \quad (2.4) $$

where $\Delta \lambda$ is the spectral bandwidth at FWHM, and $\lambda$ is the center of the wavelength of output spectrum. The pulse width is given as;

$$ Pulse \ width \ (PW) = \frac{T_{BL}}{BW} \quad (2.5) $$

From both equations 2.4 and 2.5, it is obtained that the pulse width can also be estimated from a given optical bandwidth. Generally, the pulse width of mode-locking pulses is usually measured by using an auto-correlator, which its function according to
the estimated pulse shape. The pulse shape consists of Gaussian, and Secant hyperbolic, depending on the output spectrum, characteristic of the mode-locking operation, and total cavity dispersion. The Gaussian pulse shape is obtained when the cavity dispersion is closed to zero or equal to zero as in a stretched pulse laser. Table 2.2 compares the output pulse train and an ideal TBP for two types of pulse shapes.

Table 2.2: Comparison of two types of pulse shapes

<table>
<thead>
<tr>
<th>Pulse shape</th>
<th>$I(t)$</th>
<th>$T_{BL}$</th>
<th>Time bandwidth product</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gaussian</td>
<td>$\exp(-\frac{6ln t^2}{\Delta t^2})$</td>
<td>0.441</td>
<td></td>
</tr>
<tr>
<td>Secant hyperbolic</td>
<td>$\text{sech}^2(\frac{1.76 t}{\Delta t})$</td>
<td>0.315</td>
<td></td>
</tr>
</tbody>
</table>

Passively mode-locked fiber lasers are widely explored in recent years by utilizing saturable absorption of optical materials. Various SAs have been demonstrated so far such as semiconductor saturable absorption mirror (SESAMs) (L. Sun et al., 2010), carbon nanotubes (CNTs) (Solodyankin et al., 2008), and graphene (Z. Sun et al., 2010). Mode-locking pulses have also been demonstrated in the various fiber laser cavities (Solodyankin et al., 2008; Z. Sun et al., 2010). Other nanomaterials such as MoS$_2$ and BP were also used for mode-locking pulses generation. For instance, Wang et al. demonstrated tunable mode-locked fiber laser that operates at 1.5 µm using MoS$_2$, which was obtained through liquid phase exfoliation (K. Wang et al., 2013). By using a double clad fiber, MoS$_2$ based (Rusdi et al., 2016) and BP based (Hisyam et al., 2017) were also demonstrated. Table 2.3 summarizes the performance of YDFL, which was demonstrated using various active fibers and nanomaterials SA as a mode-locker. The maximum pulse energy of the mode-locked lasers was about 32 nJ, which was obtained by using Sb$_2$Te$_3$ topological insulator materials as SA (Kowalczyk, Boguslawski, et al., 2016). The highest
repetition rate and shortest pulse duration were 21.5 MHz (Li et al., 2013) and 0.272 ps (Su et al., 2016) respectively.

Table 2.3: Mode-locked Fiber Lasers by nanomaterials based SAs

<table>
<thead>
<tr>
<th>Gain fiber</th>
<th>Materials</th>
<th>Pulse energy (nJ)</th>
<th>Pulse duration (ps)</th>
<th>Repetition rate (MHz)</th>
<th>λ (nm)</th>
<th>ΔT (%)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>YDF</td>
<td>CNTs</td>
<td>-</td>
<td>310 ps</td>
<td>21.5</td>
<td>1060</td>
<td>-</td>
<td>(Li et al., 2013)</td>
</tr>
<tr>
<td>YDF</td>
<td>Graphene</td>
<td>0:41</td>
<td>580ps</td>
<td>0.9</td>
<td>1069.8</td>
<td>8</td>
<td>(L. Zhao et al., 2010)</td>
</tr>
<tr>
<td>YDF</td>
<td>MoS$_2$</td>
<td>3.1</td>
<td>656 ps</td>
<td>6.74</td>
<td>1042.6</td>
<td>10.47</td>
<td>(Du et al., 2014)</td>
</tr>
<tr>
<td>YDF</td>
<td>MoS$_2$</td>
<td>-</td>
<td>800ps</td>
<td>6.566</td>
<td>1054.3</td>
<td>9.3</td>
<td>(H. Zhang et al., 2014)</td>
</tr>
<tr>
<td>Yb, Lu: CALGO</td>
<td>BP</td>
<td>6.48</td>
<td>0.272ps</td>
<td>63.3</td>
<td>1053.4</td>
<td>10.95</td>
<td>(Su et al., 2016)</td>
</tr>
<tr>
<td>YDF</td>
<td>BP (Double-clad)</td>
<td>5.93</td>
<td>7.54 ps</td>
<td>13.5</td>
<td>1085.5</td>
<td>8</td>
<td>(Hisyam et al., 2017)</td>
</tr>
<tr>
<td>YDF</td>
<td>Bi$_2$Te$_3$</td>
<td>-</td>
<td>960ps</td>
<td>1.11</td>
<td>1064.4</td>
<td>19.1</td>
<td>(Yan et al., 2015)</td>
</tr>
<tr>
<td>YDF</td>
<td>Bi$_2$Te$_3$</td>
<td>0.599</td>
<td>230 ps</td>
<td>1.44</td>
<td>1057.8</td>
<td>1.8</td>
<td>(Junsu Lee et al., 2015)</td>
</tr>
<tr>
<td>YDF</td>
<td>Sb$_2$Te$_3$</td>
<td>32</td>
<td>0.38 ps</td>
<td>17.07</td>
<td>1039.4</td>
<td>-</td>
<td>(Kowalczyk, Boguslawski, et al., 2016)</td>
</tr>
</tbody>
</table>

2.4 Optical Fiber Nonlinearity

The function of optical nonlinearity uses in many photonics applications. The materials have a linear reaction when the light intensity is low, which the light properties of
amplitude, polarization and phase may be modified, however no generation of new frequency component. Comparing with linear, the nonlinear response of materials obtains when the intensity of incident light is high, and that have numerous origins of nonlinear optics. There are two famous affection of nonlinearity in the materials, which are, Kerr effect and saturable absorption. Kerr effect has no energy changing between the materials and light, which is an effective model of elastic nonlinearity, associated to a harmonic motion of electrons and mostly under a light of intensive electromagnetic field. On the other hand, the linear and nonlinear reactions under electrical field ($E$) can be used to describe the overall polarization ($P$) as follows (Boyd, 2003);

$$P = \varepsilon_0 (X^{(1)} \cdot E + X^{(2)} \cdot E + X^{(3)} + \ldots)$$  \hspace{1cm} (2.6)

where $\varepsilon_0$, $X^{(1)}$, $X^{(2)}$, $X^{(3)}$ represents the vacuum permittivity, linear response of material (first order), two frequency effects (second harmonic generation), and three frequency effects (four-wave mixing), respectively.

The refractive index, $n$ under highly intensity $I$ in optical fiber is given by;

$$n = n_2 I + n_0$$  \hspace{1cm} (2.7)

where, $n_2$ is the parameter of nonlinearity that mainly used to depict Kerr effect, and $n_0$ is the linear refractive index. Saturable absorption involves an energy exchange in between light and materials and it is considered as an inelastic nonlinear effect. The nonlinear absorption of the materials under the field of light is normally related to the imaginary part of $X^{(3)}$.

When a light beam shines on a nanomaterial, electrons in the valence band are excited to the conduction band. Then these electrons gradually lose their energy due to various scattering effects and fall down to a lower position in the conduction band. When there
are many electrons excited at the same time by a highly intense light beam, the conduction band will be completely filled with electrons and no more photons will be absorbed. In this case, the absorption of the material is reduced and the material is known as “saturated” or bleached”. Such a material is called a saturable absorber (SA). The word “bleached” is used because early saturable absorbers are mostly dyes.

The nonlinear absorption, $a(I)$ according to a sample two-level SA equation is given by (Garmire, 2000);

$$a = \frac{a_s}{1 + I/I_{sat}} + a_{ns}$$  \hspace{1cm} (2.8)

where $a_s$ is the modulation depth, $I$ is the input intensity, $I_{sat}$ is the saturation intensity, and $a_{ns}$ is the non-saturable absorption. When the light intensity is low, the absorption is equal to $a_s + a_{ns}$ and when the intensity is high, the absorption is equal to $a_{ns}$. Two photon absorption (TPA) is also a nonlinear absorption effect in the materials which means the electrons absorb two photons simultaneously to be excited to the conduction band. When the light intensity is very high, TPA becomes significant enough and should be taken into consideration. The saturable model of TPA effect (J. Wang et al., 2010);

$$a = \frac{a_s}{1 + I/I_{sat}} + a_{ns} + \beta I$$  \hspace{1cm} (2.9)

where $\beta$ is the TPA coefficient. The transmission $T$ is also given by:

$$T = \exp(-a) = \exp(-\frac{a_s}{1 + I/I_{sat}} - a_{ns} - \beta I)$$

$$\approx \left(1 - \beta I - \frac{a_s}{1 + I/I_{sat}}\right)\exp(-a_{ns})$$  \hspace{1cm} (2.10)
Sometimes Equations (2.8) – (2.10) do not function admirably with the measuring data due to the requirement term \( \frac{1}{1+I/I_{sat}} \) that needs a wide transition region in order to change the state from unsaturated to saturated state.

Several researchers reported about the nonlinear measurement of saturable absorption that the transition region is narrower than the model. Therefore, the following modified model is widely used (Z. Tian et al., 2015);

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

(2.11)

In general, the two measurement methods (Z-scan and two arm measurement) are normally used to characterize the Kerr effect and saturable absorption of nanomaterials. There are two measurement techniques to characterize the Kerr effect and saturable absorption of nanomaterial. The Z-scan technique is used to measure the nonlinear absorption and nonlinear refractive index. In the Z-scan technique, the material is converted along the Z direction through the beam waist of a focused Gaussian laser beam in a narrow focusing configuration. Thus, when the material passes through the focus, the irradiance drops along with the nonlinear effects (Sheik-Bahae et al., 1990). The two-arm measurement setup of nonlinear absorption is also reported to get the modulation depth of the material (Z. Tian et al., 2015). It has a simple configuration and easier to determine the absorption of the material. The two-arm measurement setup of nonlinear absorption is shown in Figure 2.5. The mode-locked pulse from laser system is amplified using an optical amplifier to obtain a high gain peak power output to efficiently saturate the SA sample. The amplified output is connected to a variable optical attenuator before it is divided into two by a 50:50 optical coupler. The first optical power meter is linked to one port of coupler and the second port of coupler is linked to the SA sample and then the second optical power meter. The power dependent absorption measurement of the sample
can be obtained by changing the input laser source. The absorption curve can be fitted with Equation (2.11) to estimate the modulation depth and other nonlinear parameters of the SA sample.

![Two-arm measurement setup of nonlinear absorption](image)

**Figure 2.5: Two-arm measurement setup of nonlinear absorption**

### 2.5 Saturable Absorber Device

A saturable absorber (SA) is a device to modulate an optical loss inside the cavity so that it can function as a Q-switcher or mode-locker. Generally, SA effect can be realized by two approaches. The first technique uses real SAs (materials) that make the nonlinear effects reduce in absorption level and increase the light intensity. The second technique uses artificial SAs (devices), and that produce nonlinear effects into mimic the action of a real saturable absorber by inducing an intensity-dependent transmission.

The comprising of SA into the fiber laser cavity can generate pulsation either Q-switching or mode-locked, while the output properties depend on the laser cavity design and SA properties. Figure 2.6 shows the development historical of SA technologies. The first SA was a dye (Ippen et al., 1972) and the development of SA technology was slowly
evolved. A stable mode-locked pulses train was realized by using semiconductor saturable absorber mirror (SESAM) in the early 1990s (Keller et al., 1992; Zirngibl et al., 1991). Consequently, SESAM has become a successful method as SA for generation Q-switched and ultrafast mode-locked pulses from fiber lasers (L. Zhang et al., 2010). Nevertheless, its application is still limited due to their difficult fabrication methods, complex design, expensive cost and narrower wavelength operation range. These limitations are leading studies into new materials such as nanomaterials where their size that decreased dimensionally results in sturdy quantum confinement and extraordinary optoelectronic properties (Eda et al., 2013; Novoselov et al., 2005).

Figure 2.6: The evolution of SA technologies
In 1997, the first pulse generation using quantum-dot doped glasses (QD) as saturable absorbers was demonstrated (Guerreiro et al., 1997). After few years, new nanomaterials such as 1D carbon nanotubes (CNTs) and 2D graphene had drawn the great attention for pulses generation in various fiber laser setups (Bao et al., 2009; Set et al., 2004). Besides graphene, other 2D family materials that can be extracted as monolayer and few-layer crystals from a variety of bulk materials, such as topological insulators (TIs), transition metal dichalcogenides (TMDs) and black phosphorous (BP) are also gained interests (Eda et al., 2013; Kambe et al., 2014; Novoselov et al., 2005; Xia et al., 2014).

More recently, transition metal oxide material was also explored as a SA for generation Q-switching and mode-locking pulses in fiber laser (H Ahmad, Siti Aisyah Reduan, et al., 2016; Mao et al., 2017; Nady, Ahmed, Latiff, Ooi, et al., 2017). In this section, a number of new nanomaterials are introduced. These materials will be explored and used in this work for Q-switcher and mode-locker in YDFL cavity.

2.5.1 Transition Metal Dichalcogenides (TMDs)

Recently, 2D materials in layers form are building blocks for the next generation photonics technology, due to their planar quantum confinement and absence of inter-layer interactions leading to make more functional and flexible photonic devices such as graphene and transition metal dichalcogenides (TMDs). TMDs made of sandwiched metal atoms between two layers of chalcogen atoms, are the most used in electronic and optics devices owing to their direct band gap (Conley et al., 2013). Molybdenum disulfide (MoS$_2$) is one of the TMDs and its crystal structure is shown in Figure 2.7. It has 1.29 eV of band gap and good absorption coefficient of the light compared with graphene which has zero band gap. Due to this property, it is widely utilized in microelectronics devices (Wen et al., 2017) and as a catalyst (Voiry et al., 2013).
Figure 2.7: Crystal structure of MoS$_2$.

Figure 2.8: (a) Raman spectra (b) PL spectra of monolayer (1L), bilayer (2L) and trilayer (3L) MoS$_2$ sheets (Y. H. Lee et al., 2012)

Exfoliation of MoS$_2$ layers can be achieved mechanically to produce high quality nanosheets (H. Li et al., 2014) and ultrasonically in liquid phase (Jawaid et al., 2015). The liquid phase exfoliation is the most promising method to produce 2D materials in large scale. Figure 2.8 shows the Raman spectra and photo-luminescent emissions for a high quality of MoS$_2$ which were prepared using chemical vapor deposition. Raman characteristic bands around 403.8 and 385.8 cm$^{-1}$ that increase with increasing of the layers of MoS$_2$ exfoliation while PL spectra decrease with increasing in the layers of
MoS₂ exfoliation which shows two peaks in a visible range around 627 and 677 nm (Y. H. Lee et al., 2012).

2.5.2 Black Phosphorous

Another interesting 2D nanomaterial is black phosphorus (BP). It has thin atomic layers of phosphorene sheets, as shown in Figure 2.9 to produce an excellent optical and electrical properties. The band gap of a monolayer of BP is 1.88 eV and it is widely used in electronic and optoelectronic devices due to its direct band gap and excellent conductivity properties (Han Liu, Yuchen Du, Yexin Deng, & D Ye Peide, 2015). BP exfoliated layers can be produced by mechanical exfoliation using scotch-tape-based micro-cleavage and by liquid exfoliation. Scotch-tape-based micro-cleavage exfoliates mechanically the black phosphorus layers on scotch-tape and then it is transferred on substrate finally cleaned and heated to 180°C to remove the rest solvent, while liquid exfoliation is used to exfoliate the black phosphorus layer by sonicating the bulk black phosphorus powder in organic liquid. High mobility and stability of BP is normally achieved using scotch-tape-based micro-cleavage method (X. Chen et al., 2015).

![Figure 2.9: Crystal structure of few-layer black phosphorus](image-url)
Black phosphorus has three peaks for Raman spectra around 362.1, 439.5 and 467.7 cm\(^{-1}\) and this peaks remain at the same crystalline of this material with increasing in the material layers while appears one peak for photo-luminescence (PL) spectra for few layers at around 1.6 eV of photon energy, as shown in Figure 2.10 (Castellanos-Gomez et al., 2014).

2.5.3 Topological Insulator

Topological insulator (TI) materials are new kind of electronic state of matter, which have to conduct states on the surface so the electrons can only move along the surface of the materials, and are an insulator in its interior that can support the conductive surface states. Three main compounds of TI materials are Bismuth (III) Selenide (\(\text{Bi}_2\text{Se}_3\)), Bismuth (III) Telluride (\(\text{Bi}_2\text{Te}_3\)), and Antimony telluride (\(\text{Sb}_2\text{Te}_3\)).
The Bi$_2$Se$_3$ and Bi$_2$Te$_3$ are binary chalcogenides of bismuth Bi$_2$X$_3$ with complex layer structures, as shown in Figure 2.11, the crystal structure of Bi$_2$Se$_3$ and Bi$_2$Te$_3$, and directly react with the elements at 500-900°C. Bi$_2$Se$_3$ and Bi$_2$Te$_3$ have small band gaps about 1.35 and 1.21 eV, respectively, so they exhibit semiconductor properties. They can be used in television cameras, optoelectronic and switching devices, and thermoelectric refrigerator due to their properties as thermoelectric and semiconducting materials. These materials can be synthesized by physical or chemical methods (Kong et al., 2010). Chemical methods are widely used to prepare thin film of TI materials due to easy fabrication and large surface deposition (Anwar et al., 2014).

For structure mode of TI materials, the vibration modes were detected in Raman spectra as reported by Liu et al. (X. Liu et al., 2011), which are three peaks for Bi$_2$Se$_3$ and two peaks for Bi$_2$Te$_3$ at small range from 50-200 cm$^{-1}$, as shown in Figures 2.12.
2.5.4 Transition Metal Oxide

Metal oxide nanoparticles are particles contained oxygen and metal in 1 to 100 nm in size so have a high surfacing to volume ratio. They can be used to optimize light absorption and to enhance the stiffness, toughness, and probably the service life of polymeric materials. Among of these nanoparticles, nickel oxide (NiO) nanoparticles, which the crystal structure of cubic NiO is shown in Figure 2.13, is high capacitance, low cost, high chemical/thermal stability and environmental friendly. NiO nanoparticles are used as electrode material in supercapacitor, fuel cells, batteries, electrochromic films, dye-sensitized photocathodes and organic solar cells due to their catalytic, magnetic, and conductivity properties (J. Cheng et al., 2015; Kwon et al., 2016; Rai et al., 2013).
There are several methods to synthesize NiO nanoparticles such as hydrothermal, sonochemical, solvothermal, sol–gel, pulsed laser deposition, thermal decomposition and anodic plasma (Fasaki et al., 2012; Santhoshkumar et al., 2016; Zorkipli et al., 2016). To produce a thin film without using any surfactants and complex process, sonochemical method is used to synthesize nanostructure depending on the nucleation, growth and tiny bubbles collapse violently in a liquid medium. Thermal treatment is done to calcine the nanostructures into solid state which appear in different sizes with different calcination temperatures.

Cobalt (II, III) oxide (Co$_3$O$_4$) is also a transition metal oxide and a p-type semiconductor. It is a black antiferromagnetic solid and has two valence Co ions, as a mixed valance compound, Co$^{II}$ and Co$^{III}$. The Co$^{II}$ is a magnetic and occupies 1/8 of the tetrahedral sites while Co$^{III}$ is a diamagnetic and occupies ½ of the octahedral sites of the cubic close-packed lattice of the oxide anions and its formula is sometimes written as Co$^{II}$Co$^{III}_{2}$O$_4$, as shown the crystal structure of Co$_3$O$_4$ in Figure 2.14 (Heinz et al., 2013; Singh et al., 2016). Due to the owning the nanostructures unique electrical, optical,
magnetic and catalytic properties as compared to the bulk structure, Co$_3$O$_4$ nanocubes are recently a promising candidate for the application in a sensor, electrochemical devices, Li-ion batteries and photocatalysis.

![Figure 2.14: Crystal structure of Co$_3$O$_4$](image)

There are various techniques to synthesis cobalt oxide nanostructures, such as microwave assisted solvothermal, thermal decomposition and hydrothermal techniques (He et al., 2004; Numan et al., 2016; C. Sun et al., 2011). Hydrothermal method is widely used for the synthesis of Co$_3$O$_4$ nanocubes which is generally based on wet chemical route and defined as a crystal growth under various synthesis parameters such as temperature, time, pressure and concentration of precursors (Rahman et al., 2009). The structure property of the Co$_3$O$_4$ nanocubes is obtained by TEM image and XRD spectra. Feng has reported the synthesis of the Co$_3$O$_4$ nanocubes via hydrothermal process at low temperature around 160°C that is presented in TEM images in Figure 2.15 (Feng et al., 2014). Kang and Zhou has characterized a high crystallization in the cubic crystal system of the Co$_3$O$_4$ nanocubes that appeared in the XRD spectra without any noticeable
impurities, as shown in Figure 2.16 with JCPDS 42-1467 patterns, via using hydrothermal process (Kang et al., 2015).

![TEM images of Co$_3$O$_4$ nanocubes synthesized via hydrothermal process at (a) 150°C (b) 160°C (Feng et al., 2014)](image)

**Figure 2.15**: TEM images of Co$_3$O$_4$ nanocubes synthesized via hydrothermal process at (a) 150°C (b) 160°C (Feng et al., 2014)

![XRD patterns of (a) JCPDS 42-1467 and (b) Co$_3$O$_4$ nanocubes (Kang et al., 2015)](image)

**Figure 2.16**: XRD patterns of (a) JCPDS 42-1467 and (b) Co$_3$O$_4$ nanocubes (Kang et al., 2015)

The structure property of the Co$_3$O$_4$ nanocubes is characterized via Raman spectroscopy. Five peaks were observed in the Raman spectrum which is corresponded
to three $F_{2g}$, one $E_g$ and one $A_{1g}$ of the Raman active modes for the Co$_3$O$_4$ nanocrystal as marked in Figure 2.17 (Feng et al., 2014).

![Figure 2.17: Raman spectrum of Co$_3$O$_4$ nanocubes (Feng et al., 2014).](image)

2.6 Fabrication of SA

Major efforts have been considered to synthesis of controllable, uniformity and large scale of nanomaterials layer using different top-down and bottom-up fabrication approaches, including mechanical exfoliation, chemical exfoliation and chemical vapor deposition (CVD) (Johal, 2013; HF Liu et al., 2015; Yuan et al., 2016). Prior to the preparation of the nanomaterials layer, there are different methods that can prepare the nanomaterials in the nanoscale from the bulk materials mainly using physical and chemical methods such as hydrothermal, sonochemical, solvothermal, sol–gel, pulsed laser deposition and thermal decomposition (Alagiri et al., 2012; Aslani et al., 2011; Fasaki et al., 2012; X. Liu et al., 2005; Sietsma et al., 2007). Here, the fabrication methods are briefly described.
2.6.1 Mechanical Exfoliation Techniques

Nanomaterials can be prepared by using an exfoliation of the bulk materials which can be peeled into a layer by layer. That is due to the weak bonding between atoms mainly van der Waals bond which the intermolecular force is weak. Therefore, the material crystal is easy to dissociate between layers under the external force. This method is preferred for 2D nanomaterials fabrication due to low cost, easy and less time fabrication and produces high quality and a large area of the 2D nanomaterials. The graphene and TI layers can be prepared by stripping the graphite and TI bulk materials. Hong et al. reported the preparation of Bi$_2$Se$_3$ nanoribbons by exfoliation down of the bulk material to a few quintuple layers (Hong et al., 2010). Graphene was prepared by mechanical exfoliation using scotch-tape–based micromechanical cleavage (Yi et al., 2015). The process of this method is shown in Figure 2.18.

![Figure 2.18: An illustrative procedure of the Scotch-tape–based micromechanical cleavage for graphene (Yi et al., 2015)](image-url)
2.6.2 Liquid Phase Exfoliation Technique

2D nanomaterials can also be prepared using liquid phase exfoliation (LPE) approach, which is based on breaking layered materials into 2D nanomaterials. A chosen solvent is added to the layered materials that are easy to suspend and break the material crystal in the solvent. The LPE process depends on three general steps, dispersion the bulk material into the chosen solvent, sonication then centrifugation. The propagation of high amplitude sonication waves in the sonication process induces the shear forces and cavitation in the solution leading to shearing the crystal. Recently, this method is widely used for exfoliation layered materials such as black phosphorous and transition metal dichalcogenides (Gupta et al., 2016; S. Lin et al., 2017). Cui et al. has successfully fabricated the graphene using solvothermal-assisted exfoliation process in acetonitrile (ACN) solvent, with the production of the graphene nanosheets (GNS) (Cui et al., 2011). The detailed mechanism of the solvothermal exfoliation processes of graphene is schematically illustrated in Figure 2.19.

Figure 2.19: Solvothermal exfoliation processes of graphene (Cui et al., 2011)
2.6.3 Sonochemical Technique

Sonochemical technique is one of the promising method to synthesis nanomaterials especially transition metal oxide nanoparticles due to not using any surfactant and complex process (Kis-Csitári et al., 2008; Kumar et al., 2000). The sonochemical is considered the effect of ultrasound in forming acoustic cavitation in liquids that results in the enhancement of the chemical reaction in the solution mainly the nucleation, growth and tiny bubbles. Mousavi et al reported about the synthesis of Tellurium (Te) nanoparticles via ultrasonics sonochemical method while Te nanorods were produced in the absence of ultrasonic waves that is illustrated in Figure 2.20 (Mousavi-Kamazani et al., 2017). In addition, NiO nanostructure was successfully synthesized using facile sonochemical method following by calcination process. The NiO nanoclusters were formed at 250 °C of calcination temperature (Duraisamy et al., 2016).

![Te nanoparticle and Te nanorod](image)

**Figure 2.20:** Schematic diagram of the formation of Te nanoparticles and effect of ultrasonic irradiation on the particle size and morphology (Mousavi-Kamazani et al., 2017)

2.6.4 Other Techniques

In addition to the above techniques, there are other fabrication techniques. In 2014, Liu et al had synthesized a high quality of Bi$_2$Se$_3$ layers using chemical vapor deposition (CVD) system using high purity Se and Bi$_2$Se$_3$ powder as source materials (M. Liu et al.,
2014). Bi₂Te₃ thin film on GaAs substrate was fabricated using a new technology called Metal-organic chemical vapor deposition (MOCVD). Moreover, the pressure and temperature are important in the MOCVD technique that leads to increase in the mobility of Bi₂Te₃ at 7400 cm²/V.s at 15 K from the bulk carrier mobility (350 cm²/V.s) at 30 K (Cao et al., 2012).
CHAPTER 3: MOLYBDENUM DISULFIDE AS SATURABLE ABSORBER

3.1 Introduction

Fiber lasers use trivalent rare-earth ions such as ytterbium, erbium, and thulium, which was doped into glass hosts as a gain medium. At 1962, the first laser performance based on ytterbium-doped silicate fiber was investigated. Then, the Yb$^{3+}$ has gained a widespread interest as a gain medium to replace the conventional inefficient Nd$^{3+}$. This is attributed to the Nd$^{3+}$ which operates based on four level transition, while Yb$^{3+}$ has only three level and quasi-three level transitions. The ytterbium-doped fiber (YDF) is attractive as a gain medium because it has a simple energy level structure, high reliability, compact and the lasing wavelength at 1 µm, within the low-loss of optical laser as well as it is capable for producing high output power.

Q-switched and mode-locked lasers with the advantages of compactness and flexibility, have attracted much attention in recent years due to their potential applications in laser material processing, remote sensing, telecommunication and medicine (Bao et al., 2011; S. Harun et al., 2012; Ursula Keller et al., 1996). Pulsed YDF lasers (YDFLs) operating in 1 micron region have been widely investigated due to their excellent efficiency (Harith Ahmad et al., 2016; Akulov et al., 2007; Al-Masoodi et al., 2014). They are normally obtained using active approaches by inserting an acoustic-optic or an electron-optic modulator into the laser cavity. Recently, passive methods are also widely explored for both Q-switching and mode-locking in fiber lasers by utilizing the saturable absorption of optical materials (i.e. saturable absorbers, SAs). In the past, several kinds of SAs such as semiconductor saturable absorber mirrors (SESAMs) (Ge et al., 2009; X. Wang et al., 2010), carbon nanotubes (CNTs) (Garnov et al., 2007; Zhou et al., 2010), graphene (J Lee et al., 2013) have been successfully used to passively generate both Q-switched and mode-locked fiber lasers. Although SESAMs have been well developed and commercially available for the past two decades, they exhibit obvious disadvantages such
as complex fabrication, expensive packaging and low damage threshold. Graphene and carbon materials have the advantages of low cost, easy fabrication and operable in a wide wavelength range. Mode-locking pulses have been demonstrated using graphene and carbon nanotubes (CNTs) as SA in the various fiber laser cavities based on erbium- (Mou et al., 2012; Z. Sun et al., 2012; Xu et al., 2012), ytterbium- (H.-R. Chen et al., 2014; X.-H. Li et al., 2012; L. Zhao et al., 2010) and thulium- (Cho et al., 2009; H. Zhang et al., 2009) doped fiber gain medium. The operation wavelength of CNT is related with diameter of the nanotubes. Graphene has a 2D zero-bandgap semiconductor material and low absorption coefficient, which gives it the advantages of wideband saturable absorption as well as ultra-fast recovery time. These advantages lead researchers to 2D materials.

In this chapter, molybdenum disulfide (MoS$_2$) is proposed as SA for generating Q-switched and mode-locked pulses train in YDFL cavity. A passively Q-switched YDFL is demonstrated using a mechanically exfoliated from a natural MoS$_2$ crystal. The few layer MoS$_2$ is obtained by using a scotch tape, which is then sandwiched between two fiber ferrules to form a fiber compatible Q-switcher. By introducing the MoS$_2$ SA into the YDFL cavity, a stable pulse laser is generated at 1070.2 nm wavelength with repetition rate is tunable from 3.817 to 25.25 kHz. A mode-locked YDFL is also realized using another MoS$_2$ based SA, which was realized by embedding the MoS$_2$ into a polymer film. The mode-locked laser cavity is constructed based on a 10.6 m long cavity with normal dispersion of 0.242 ps$^2$/km.nm at 1030 nm.

### 3.2 Passively Q-switched Ytterbium Doped Fiber Laser with Mechanically Exfoliated MoS$_2$ Saturable Absorber

Compact and flexible passively Q-switched fiber lasers have opened up new applications in laser material processing, remote sensing, telecommunications and
One of the most effective methods for realizing a Q-switching pulse train in fiber laser cavity is through the use of a SA device as a Q-switcher. In the past, various SAs namely semiconductor saturable absorber mirror SESAM (Hakulinen et al., 2007), carbon nanotubes CNTs (S. Harun et al., 2012; Li et al., 2016) and graphene (Al-Masoodi et al., 2015; Ismail et al., 2013), have been successfully used to passively generate Q-switched fiber lasers. Even today, researchers are still searching for new SAs which possess desirable characteristics such as wavelength-independent saturable absorption, low saturable optical intensity, high damage threshold, and excellent fiber compatibility.

More recently, Molybdenum Disulfide (MoS$_2$) has also been tested as an SA for mode-locking and Q-switching (Dong et al., 2015; Z. Lin et al., 2016; H. Liu, A.-P. Luo, et al., 2014; Z. Luo et al., 2014; K. Wang et al., 2013; R. Woodward et al., 2014; Zhan et al., 2015; H. Zhang et al., 2014). For instance, Wang et al. investigated the saturable absorption property of two-dimensional (2D) MoS$_2$ nanosheets for ultrafast laser applications (K. Wang et al., 2013). Other reported works indicate that MoS$_2$ has a higher modulation depth than that of graphene (~2.3% per layer) (H. Cheng et al., 2016; Tang et al., 2013; K. Wang et al., 2014; S. Zhang et al., 2015). In this section, a new passively Q-switched YDFL is demonstrated using a newly developed MoS$_2$ based SA as a SA. The MoS$_2$ SA was prepared by mechanically exfoliating the crystal using a scotch tape and sandwiching the tape with acquired flakes between two fiber ferrules using a fiber connector. The laser operates at 1070.2 nm with a high pulse energy of 295.45 nJ. The proposed laser produces a higher pulse energy compared to the previous works on MoS$_2$ based Q-switched fiber lasers, which so far only reported the maximum pulse energy of 126 nJ (Z. Luo et al., 2014; R. Woodward et al., 2014). The laser also performs competitively compared to similar lasers in 1 µm region that use other 2D materials as SAs (X. Li et al., 2014; Tang et al., 2013).
3.2.1 Fabrication and Characterization of Mechanically Exfoliated MoS$_2$ based SA

MoS$_2$ is a nanomaterial which has extraordinary electronic and optical properties (Mak et al., 2010). The MoS$_2$ layers used in the construction of the SA were obtained by mechanical exfoliation method, which was performed without using any chemical substance and costly instrument. The fabrication process is summarized in Figure 3.1. Figure 3.1 (a) shows the relatively thin flakes that was evenly peeled off a natural MoS$_2$ crystal by scotch tape. The flakes attached to the scotch tape was repeatedly pressed to make it thin enough to transmit light with a high efficiency. Then a tiny piece of the MoS$_2$ tape was cut and placed onto the end face of a FC/PC fiber connector ferrule after applying index matching gel as shown in Figure 3.1 (b). After connecting it with another FC/PC ferrule with a standard flange adapter, the all-fiber MoS$_2$ SA was finally completed and ready to be used. It was learned that if a clean scotch tape is placed onto the end face of fiber connector, the cavity will fail to generate Q-switching regime.

![Figure 3.1: Mechanical exfoliation method; (a) Simple peeling process, and (b) MoS$_2$ tape at standard FC/PC connector ferrule end surface](image-url)
Figure 3.1, continued

Raman spectroscopy was then performed on the fabricated MoS$_2$ SA. Figure 3.2 (a) shows the Raman spectrum recorded by a spectrometer when a 514 nm beam of an Argon laser was radiated on the MoS$_2$ tape for 10 seconds with an exposure power of 10 mW. As shown in the figure, the sample exhibits two characteristic peaks in parallel with two phonon modes; out of plane vibration of Sulfide atoms at 408 cm$^{-1}$, and in plane vibration of Molybdenum and Sulfide atoms at 383 cm$^{-1}$ with a frequency difference of 25 cm$^{-1}$. This frequency difference indicates that the prepared tape contains the layers of MoS$_2$ (H. Li et al., 2012). Following other reported results (Wu et al., 2015), the full-width-half-maximum (FWHM) of $E_{2g}^{1}$ and $A_{1g}$ bands were calculated to be 5.0 and 5.5 cm$^{-1}$, respectively. Figure 3.2 (b) shows the linear absorption of the SA within a wavelength range from 1520 to 1600 nm. It was obtained by transmitting a broadband light source through the MoS$_2$ tape. As shown in the figure, the absorption follows a flat profile across the wavelength region. The inset of Figure 3.2 (b) shows the absorption spectra within UV-Vis region. From the figure, two exciton peaks of MoS$_2$ are noticeable at 612 nm and 670 nm. These peaks correspond to the interband excitonics transition at 2.03 eV and 1.85 eV, respectively. The absorption spectra obtained are comparable to those found in the previous studies (M. H. M. Ahmed et al., 2016).
The nonlinear optical response property of the MoS$_2$-SA tape was further investigated to confirm its saturable absorption. This was done by applying a balanced twin-detector measurement technique. A self-constructed mode-locked fiber laser (1557 nm wavelength, 1.5 ps pulse width, 17.4 MHz repetition rate) was used as the input pulse source. As in the work of Fu et al., a mode-locked laser source was launched to generate lasers at 1, 1.5 and 2 µm (Fu et al., 2014). In this work, the transmitted power was recorded as a function of incident intensity on the device by varying the input laser power. The experimental data for transmission were fitted to a simple two-level SA model:

\[ T(I) = 1 - \alpha_s \exp \left( -I/I_{sat} \right) - \alpha_{ns} \] (3.1)

where \( T(I) \) is the transmission, \( \alpha_s \) is the modulation depth, \( I \) is the input intensity, \( I_{sat} \) is the saturation intensity, and \( \alpha_{ns} \) is the non-saturable absorption. The nonlinear transmission of the MoS$_2$ on the scotch tape is shown in Figure 3.2 (c). As seen, the modulation depth, non-saturable intensity, and saturation intensity are 11.3 %, 23.0 %, and 23.5 MW/cm$^2$, respectively.

![Raman Spectrum](image1)

**Figure 3.2:** Optical characteristic of the MoS$_2$ tape. (a) Raman spectrum. (b) Linear absorption. Inset image is absorption at UV-Vis region. (c) Nonlinear transmission.
Figure 3.2, continued
3.2.2 Experimental Setup

The experimental setup of the proposed MoS$_2$ based Q-switched YDFL is schematically shown in Figure 3.3. The laser cavity consists of a 980/1030 nm wavelength division multiplexer (WDM), 1.5-m long of Ytterbium-doped fiber (YDF), MoS$_2$ SA, an isolator and a 3 dB output coupler in a ring configuration. The YDF used as a gain medium, has a numerical aperture of 0.20, a core and cladding diameters of 4 and 125 µm, respectively, as well as ion absorption of 280 dB/m at 920 nm. A unidirectional operation of the laser cavity is obtained by using an isolator. The output of the YDFL is collected from the cavity via a 50:50 coupler, which retains 50% of the light in the laser ring cavity for oscillation. The 3dB coupler was used to increase the cavity loss to avoid mode-locking instability and achieve a stable Q-switching operation. The optical spectrum analyser (OSA) used for analyzing the Q-switching has a spectral resolution of 0.02 nm. The oscilloscope is used to observe the output pulse train of the Q-switched laser.

![Figure 3.3: Experimental setup for Q-switched YDF fiber ring laser using SA](image-url)
3.2.3 Q-switching Performance

The YDFL started lasing in Q-switched mode at a pump power of 49.57 mW. When the pump power was gradually increased from 49.57 mW to 87.2 mW, the repetition rate of the pulse increased in response. This is a typical feature of passive Q-switching. Figure 3.4 shows the optical spectra of the Q-switched YDFL at three different pump powers of 49.57 mW, 71.3 mW, and 87.2 mW. While the peak of the YDFL stays fixed at 1070.2 nm, its 3 dB spectral bandwidth widens with the increase in pump power. The bandwidth are 0.2 nm, 0.4 nm and 0.5 nm at the three pump powers respectively. To verify that the passive Q-switching was attributed to the MoS$_2$ SA, the MoS$_2$ tape was removed from the ring cavity. In this case, no Q-switched pulses were observed on the oscilloscope, even when the pump power was adjusted to a wide range. This finding confirms that the MoS$_2$ SA was responsible for the passively Q-switched operation of the laser.

![Optical spectra](image)

Figure 3.4: Optical spectra of the Q-switched YDFL configured with the MoS$_2$ SA at different pump power of 49.57 mW, 71.3 mW, and 87.2 mW
Figure 3.5 (a), (b) and (c) show the pulse trains and single-pulse envelops of the Q-switched YDFL at pump powers of 49.57, 71.3, and 87.2 mW, respectively. As the pump power increases, more energy can be stored in the laser cavity and this contributes to the rise in the repetition rate accompanied by the reduction in pulse width (Y. Chen et al., 2014). For pump powers of 49.57, 71.3, and 87.2 mW, the attained repetition rates are 3.817 to 17.54 and 25.25 kHz while the corresponding pulse widths are 26.09, 11.27 and 11.33 µs, respectively. The attributes of the pulse a Q-switched laser mainly depend on the gain medium, SA and pump power.
Figure 3.6 shows the repetition rate and pulse width of the Q-switched YDFL as a function of the pump power. As the pump power increases from 49.57 to 87.2 mW, the repetition rate grows from 3.817 to 25.25 kHz. Throughout the experiment, the output was stable and no amplitude modulations were observed. This indicates that there was no self-mode locking (SML) effect during the Q-switching operation. On the other hand, the pulse width decreases from 26.09 μs to 11.33 μs. As the pump power exceeds 71.3 mW, the pulse width stays nearly unchanged, which clearly indicated that the SA was saturated. It was reported that the pulse width of a passively Q-switched laser depends on the cavity round-trip time and the modulation depth of the SA (Zayhowski et al., 1991). Therefore, the minimum pulse duration obtained in our experiment can surely be narrowed by shortening the cavity length and improving the modulation depth of the MoS$_2$ layer.
Figure 3.6: Pulse repetition rate and pulse width of the proposed Q-switched YDFL versus incident pump power

Figure 3.7 shows the dependency between average output power and single pulse energy of the proposed Q-switched YDFL against the pump power. As shown, the average output power increases linearly from 0.542 to 7.46 mW while the pulse energy also increases from 142 to 295.45 nJ as the pump power rises from 49.57 to 87.2 mW. The pulse energy is calculated based on the measured average output power and the repetition rate. When the pump power goes past 87.2 mW, the Q-switched pulse train becomes unstable or disappears, as typically observed in certain passively Q-switched fiber lasers (Z. Luo et al., 2013). Subsequently, when the pump power is reduced to a level below 87.2 mW, the stable Q-switched pulse train reappear. Hence, the unstable Q-switched operation at high pump power may be attributed to over-saturation of the MoS$_2$ SA rather than to thermal damage (Z. Luo et al., 2013). Figure 3.8 shows the RF spectrum of the Q-switched YDFL at the pump power of 87.2 mW, for which the pulse repetition rate is 25.25 kHz, matching the oscilloscope data of Figure 3.5 (c). The RF signal-to-noise ratio (SNR) is about 30 dB with no spectral modulation, indicating that the passively Q-switching operation is stable.
It is worthy to note that the mode-locking pulses train was failed to be obtained with the current SA. This is most probably due to the large number of MoS$_2$ layers in the SA as well as the high loss at the scotch tape. Therefore, a new SA was developed in the next section using a liquid phase exfoliation method.

Figure 3.7: Average output power and pulse energy of the proposed Q-switched YDFL versus incident pump power

Figure 3.8: RF spectra of the Q-switched YDFL at the pump power of 87.2 mW with 150 kHz span. Inset is an enlarge image of 25.25 kHz repetition rate
3.3 Passively Mode-locked YDFL Operation with Few Layer MoS\textsubscript{2} Embedded into Polyvinyl Alcohol Composite Polymer as a SA

In the recent years, mode-locked fiber lasers have extensively engaged interest due to their application benefits, such as micromachining, communication, and optical systems; as well as their nature being compact, cost-effective and easy to setup (Grelu et al., 2012; Lecaplain et al., 2012). Active mode-locking pulse train can be achieved by inserting electronic switching such as acousto-optic modulator (Ryu et al., 2007). Passively mode-locked fiber lasers are also widely explored in recent years by utilizing saturable absorption of optical materials. Various SAs have been demonstrated so far such as SESAMs (L. Sun et al., 2010; M. Zhang et al., 2009), carbon nanotubes (CNTs) (Mou et al., 2012; L Zhang et al., 2011), and graphene (Z. Sun et al., 2012; Z. Sun et al., 2010). SESAMs are good SA device which can be used to generate a mode-locked fiber laser in both 1 μm and 1.5 μm regions (L. Chen et al., 2009; M. Zhang et al., 2009). However, its application is still limited due to their difficult fabrication methods, complex design, expensive cost and narrower wavelength operation range.

To date, another 2D nanomaterial, MoS\textsubscript{2} is also drawing more interest especially for mode-locked fiber laser applications. The reason for this growing interest is due to its material’s thickness, which gives an excellent optical laser properties as well as the band-gap (M. Ahmed, A. Latiff, H. Arof, H. Ahmad, et al., 2016; H. Liu, A.-P. Luo, et al., 2014). Until today, only a few works have been reported on MoS\textsubscript{2} based SA (M. Ahmed, A. Latiff, H. Arof, H. Ahmad, et al., 2016; Du et al., 2014; H. Liu, A.-P. Luo, et al., 2014; Sathiyan et al., 2016; M. Zhang et al., 2015). In the previous section, a Q-switched EDFL was demonstrated using a mechanically exfoliated MoS\textsubscript{2} as a Q-switcher. In this section, a passively mode-locked YDFL is demonstrated using a few layered MoS\textsubscript{2} as a SA. The few layer of MoS\textsubscript{2} polymer is a composite of liquid phase exfoliation (LPE) of chemically pristine crystal. A small piece of MoS\textsubscript{2} polymer film was inserted in the YDFL cavity,
sandwiching it in between two fiber optical ferrules with a connector to obtain mode-locking pulses with repetition rate of 18.8 MHz and pulse energy of 0.1 nJ. The LPE method is simple, cheap and does not require any complicated procedures.

### 3.3.1 Fabrication and Characterization of the MoS$_2$ Film as SA

The liquid phase exfoliation (LPE) process for MoS$_2$ exfoliation was carried out in two steps as shown in Figure 3.9. The bulk MoS$_2$ crystals were first mixed together with a solvent that gets a comparable surface energy with MoS$_2$ (~75 mJ·m$^{-2}$) (Cunningham et al., 2012). Likewise, Dimethylformamide (DMF) solvent was chosen with a concentration of 5 mg/ml for this work. Following, the second step was to exfoliate the material through a sonication process. In this process, local pressure variations were generated to sufficiently separate the atomic layers of the bulk crystal (Coleman et al., 2011; Hasan et al., 2010) in the solvent to produce a dispersion, which was rich in few-layer flakes (Coleman et al., 2011). Then, the MoS$_2$ dispersion was kept to be ultrasonicated for 24 hours before it was centrifuged for 1 hour at ~3000 rpm. This allowed the un-exfoliated sediment to settle at the bottom since the larger flakes descended more rapidly than the exfoliated few-layer flakes in the centrifuge cell (Bonaccorso et al., 2013). The top portion of the dispersion containing few-layer flakes was taken out for further composition fabrication process.

A few procedures were followed in the second step to produce MoS$_2$ polymer film for this experiment. Firstly, ~10 ml of decanted MoS$_2$ dispersion was put together with a 10 mg/ml concentrated ~20 ml of polyvinyl alcohol (PVA) solution. This solution was a result of ~200 mg of PVA being dissolved in 20 ml of deionized water. By this, 30 ml of MoS$_2$-PVA was prepared, which was then heated at a temperature of ~80 °C and stirred with a magnetic object until the solution was brought down to an approximate volume of ~10 ml. These series of actions took around 7 hours to be done, following which the
resulting MoS$_2$-PVA solution was placed in a glass substrate and put in an oven for 4 hours at a temperature of ~80 °C in order to dry and obtain a composite film with a thickness of ~40 μm.

**Figure 3.9: The fabrication process of MoS$_2$ PVA film**

Field emission scanning electron microscopy (FESEM) and Raman spectroscopy were performed on the fabricated MoS$_2$–PVA film sample. Figure 3.10 (a) shows the FESEM image that illustrates a MoS$_2$ nanoplatelets embedded with small particles in average sizes from 3-10 μm with slight bends. Figure 3.10 (b) presents the Raman spectrum recorded using a spectrometer at a 514 nm beam of an Argon-ion laser that was radiated on the film for 10 seconds with an exposure power of 10 mW. As shown in the figure, the sample displays two signature peaks at 383 cm$^{-1}$ and 407 cm$^{-1}$ which are related to in-plane vibration of Molybdenum and Sulfide atoms and out of plane vibration of Sulfide atoms respectively. The frequency difference was recorded to be 24 cm$^{-1}$. It was observed that the $E_{2g}^1$ mode according to the in-plane motion shifted to a shorter wavelength after complete exfoliation. This indicated that the number of layers of MoS$_2$ were in the range of 2-5 layers (H. Li et al., 2012).
Figure 3.10: (a) FESEM image and (b) Raman spectrum of the MoS₂-polymer composite film
A nonlinear absorption measurement was also performed on the fabricated MoS$_2$-PVA film to confirm its saturable absorption as shown in Figure 3.11 (a). In the experiment, a stable mode-locked fiber laser operating at 1565 nm wavelength with repetition rate of 15 MHz and pulse width of 0.9 ps, was employed as the input pulse source. The mode locking signal results were then amplified using an erbium-doped fiber amplifier (EDFA) in order to obtain a high peak power output to efficiently saturate the MoS$_2$ sample. The output port of the amplifier was connected with a variable optical attenuator to control the launching output power to the system before it is launched into a 3dB optical coupler and two optical power meters. The first optical power meter was linked to one port of coupler and the second port of coupler was linked to the MoS$_2$ film sample then connected into the second optical power meter. The resultant characteristic of MoS$_2$ film as SA is shown in Figure 3.11 (b).

The absorption power was recorded as a function of incident intensity on the device by modifying the input optical laser power. The experimental data for absorption were prepared depending on the two-levels of SA model (Garmire, 2000).

$$a(I) = \frac{\alpha_s}{1 + I/I_{sat}} + \alpha_{ns}$$  \hspace{1cm} (3.2)

where $a(I)$, $\alpha_s$, $I$, $I_{sat}$ and $\alpha_{ns}$ are stand for absorption, modulation depth, input intensity, saturation intensity, and non-saturation loss respectively. For the MoS$_2$ sample that was used in this work, the modulation depth, non-saturable intensity, and saturation intensity were measured to be approximately 9.7 %, 10.9 %, and 40 MW/cm$^2$, respectively.
Figure 3.11: The nonlinear absorption measurement of MoS$_2$-SA. (a) Experimental setup for nonlinear measurement. (b) Nonlinear absorption curve

3.3.2 Configuration of the Mode-Locked Laser

The proposed configuration of mode-locked YDFL with MoS$_2$ SA is shown in Figure 3.12. It uses a 0.7 m long of Ytterbium-doped fiber (YDF) with 1300 dB/m peak absorption at 977 nm as a gain medium. The YDF was pumped by a 974 nm laser diode through a 980/1060 nm wavelength division multiplexer (WDM). An isolator is used in the ring cavity to avoid backward reflection as well as ensure unidirectional operation.
The MoS$_2$ SA device was composed by sandwiching the fabricated MoS$_2$ PVA film in between two fiber ferrules via an adapter as shown in the inset of Figure 3.12. It is incorporated into the laser cavity to act as a mode-locker for generating ultrashort pulses train. A 10 dB coupler is used to tap out the output laser while allowing 90% of the photons to oscillate inside the cavity. The cavity length is measured to be about 10.6 m with normal dispersion of 0.242 ps$^2$/km.nm at 1030 nm. The output spectrum of the laser is measured by an OSA with a 0.02 nm spectral resolution while the temporal characteristic of the laser is investigated using an oscilloscope coupled with a 2-GHz photodetector. The RF spectrum is measured by an RF spectrum analyzer with a range operation from 9.0 kHz to 7.8 GHz.

![Experimental setup for mode-locked YDFL with MoS$_2$ based SA. Inset shows how the MoS$_2$ PVA film is incorporated in the laser cavity](image)

**Figure 3.12:** Experimental setup for mode-locked YDFL with MoS$_2$ based SA. Inset shows how the MoS$_2$ PVA film is incorporated in the laser cavity

### 3.3.3 Mode-locking Performance

At first, we observed a continuous wave (CW) laser operation when the pump power reached to 58.0 mW. As the pump power was further increased to 78.7 mW, the self-
started and stable mode-locked pulse train was generated. The mode-locked pulse train was maintained with further increase in pump power up to the maximum pump power of 202.9 mW. Figure 3.13 (a) shows the output spectrum of the mode-locked laser at pump power of 202.9 mW. The peak wavelength was centered at 1073.86 mW with 3dB bandwidth of 0.06 nm without Kelly sideband, which indicates that the pulses operate in normal dispersion.

The typical mode-locking pulse train at pump power of 202.9 mW is shown in Figure 3.13 (b). It is observed that the peak to peak spacing of the pulse train was about 53 ns, which equals to the repetition rate of 18.8 MHz. The repetition rate obtained is corresponded with the fundamental frequency for cavity length of 10.6 m. As shown in the figure, the pulse width is measured to be 3.12 ns, which is a lot broader than the actual pulse width. This is attributed to the oscilloscope resolution limitation. To estimate the actual pulse width, we used mathematical calculation based on time bandwidth product (TBP). Assuming that the TBP is 0.441 for Gaussian pulse profile (Hisyam et al., 2017) minimum possible pulse width is calculated to be approximately 28.3 ps.

Figure 3.13 (c) shows the output power and pulse energy of the mode-locked laser at various pump powers. As seen in the figure, both output power and pulse energy linearly increased with the pump power. At the maximum pump power of 202.9 mW, the maximum output power and pulse energy are obtained at 1.84 mW and 0.1 nJ, respectively. Figure 3.13 (d) shows the RF spectrum for YDFL laser. The fundamental frequency of the laser is obtained at 18.8 MHz with a SNR of more than 30 dB, which indicates the stability of the mode-locking operation. It is worthy to note that there is no appearance of fundamental frequency noticed as the MoS\(_2\) is taken off at all level of pump power.
Figure 3.13: Mode-locked YDFL performances. (a) Output spectrum and (b) Oscilloscope train at maximum pump power. (c) Output power and pulse energy. (d) RF spectrum

\[ \lambda = 1073.86 \text{ nm} \]
\[ \Delta \lambda = 0.06 \text{ nm} \]
Figure 3.13, continued
3.4 Summary

In this chapter, a simple and compact passive Q-switched and mode-locked YDFL was successfully demonstrated using MoS$_2$ based SA. It was found that a Q-switching operation at 1µm region can be realized by using a bulk layer of MoS$_2$ tape, which was obtained by a mechanical exfoliation technique. The tape was sandwiched between two fiber ferrules to form the fiber compatible MoS$_2$ based Q-switcher. A Q-switched pulse train operating at 1070.2 nm was obtained as the 980 nm pump power reached the threshold value of 49.57mW and it continued to operate stably until 87.2 mW. The corresponding pulse repetition rate was tunable from 3.817 to 25.25 kHz. The lowest pulse width was 11.33 µs and the maximum pulse energy was 295.45 nJ.

In the other hand, a passive mode-locked with 0.7 m long of YDF as a gain medium was successfully reported using a few layered MoS$_2$ PVA film as a mode-locker. The MoS$_2$ film was obtained based on a LPE of MoS$_2$ crystal. Stable and self-starting passive mode-locking pulses were achieved within the pump power of 78.7 mW to 202.9 mW. It operates at 1073.86 nm with a fixed repetition rate of 18.8 MHz with picoseconds pulse width. The maximum pulse energy was calculated to be approximately 0.1 nJ at pump power of 202.9 mW. This piece of work tackled an important aspect that contributes to study nonlinear optical properties of MoS$_2$, which may in turn guide to future paths for ultrafast photonics applications. These experimental results suggest that MoS$_2$ is a promising material for pulsed laser applications.
CHAPTER 4: BLACK PHOSPHORUS AS SATURABLE ABSORBER

4.1 Introduction

As discussed in the previous chapters, passively Q-switched and mode-locked fiber lasers have gained widespread attention due to their advantages in compactness and flexibility, and potential applications in medicine, telecommunications, remote sensing and laser material processing. To date, many forms of saturable absorber (SA) have been used in generating these lasers including semiconductor saturable absorber mirror (SESAM) (U. Keller et al., 1996), single-walled carbon nanotubes (SWCNTs) (S. W. Harun et al., 2012) and graphene (H.-P. Li et al., 2014; J. Zhao et al., 2014). Despite the successful implementation of these SAs, researchers are still looking for new SAs with better characteristics in terms of wavelength-independent saturable absorption, low saturable optical intensity, fiber compatibility and high damage threshold. Recently, topological insulators (TIs) and Molybdenum disulfide (MoS$_2$) have also been identified as potential SAs for mode-locking and Q-switching (Y. Chen et al., 2013; Z. Luo et al., 2013; Ren et al., 2015).

In the previous chapter, MoS$_2$ as SA was proposed and reported for generating both Q-switching and mode-locking in Ytterbium-doped fiber laser (YDFL) cavity. Another 2D material: a black Phosphorus (BP) dubbed as the newly emerging allotrope, has gained much attention for potential applications in the next-generation optoelectronics devices such as sensor, field-effect transistors and solar cells (Abbas et al., 2015; J. Dai et al., 2014; L. Li et al., 2014). Multi-layer BP has a similar structure as bulk graphite. In a single layer, each phosphorus atom is covalently bonded with three adjacent phosphorus atoms to form a stable honeycomb structure, and different layers are stacked together by van der Waals interaction (Han Liu, Yuchen Du, Yexin Deng, & Peide D. Ye, 2015). It has the common properties of 2D materials such as wideband absorption, ultrafast carrier dynamics and planar characteristic (J. Qiao et al., 2014). The BP comprises only the
elemental “phosphorus” and thus it could be easily peeled off by mechanical exfoliation. Tunable Q-switched fiber laser was demonstrated based YDFL using Black Phosphorus (BP) as SA with maximum pulse energy of 7.1 nJ (Harith Ahmad et al., 2016). In another work, dual wavelength Q-switched YDFL with maximum pulse energy of 2.09 nJ. Mode-locking pulse was also generated in double cladding YDF laser using BP as SA with repetition rate of 13.5 MHz and pulse duration of 7.54 ps (Hisyam et al., 2017).

In this chapter, Black phosphorus (BP) is proposed as new SA for operation at 1 micron region. Both Q-switching and mode-locking operations are demonstrated using a ytterbium doped fiber (YDF) as a gain medium. At first, a Q-switched YDFL is demonstrated using a newly developed multi-layer BP SA. The SA is prepared by fixing the BP material onto a sticky tape using a mechanical exfoliation technique as described in the previous chapter. The BP tape is placed between two ferrules and incorporated in YDFL cavity to achieve a stable Q-switching pulse operating at 1.0 µm region. The laser has a pump threshold of 55.1 mW, a pulse repetition rate that is tunable from 8.2 to 32.9 kHz, and the narrowest pulse width of 10.8 µs. The highest pulse energy of 328 nJ is achieved at the pump power of 97.6 mW.

In mode-locking experiment, the BP SA is prepared using a slightly different method to reduce the device loss and thus enable the mode-locking operation. The SA is prepared by mechanically exfoliation of the BP crystal and fixed the acquired flakes onto the end surface of a standard FC/PC fiber connector. By incorporating the BP SA into the YDF laser cavity, a stable mode-locked operation is obtained at 1033.76 nm wavelength and the repetition rate of 10 MHz. The maximum pulse energy and peak power are almost 2.7 nJ and 0.83 kW respectively at the maximum pump power of 200 mW. These experimental results indicate that the BP SA can be used successfully to generate stable Q-switching and mode-locking pulses train at 1 µm region.
4.2 BP SA for Q-switching

In this section, a passively Q-switched YDFL is demonstrated using multi-layer BP based SA, which was obtained by mechanical exfoliation method. Mechanical exfoliation is advantageous mainly because of its simplicity and reliability, where the entire fabrication process is free from complicated chemical procedures and costly instruments.

4.2.1 Fabrication and Standard Characterization of the BP based SA

The multi-layer BP used in our experiment was prepared by peeling bulk-structured BP 2D thin layers using a scotch tape. Here, a large block of commercially available BP crystal (with purity of 99.995 %) was used as a raw material. Then, the flakes on the scotch tape was repeatedly pressed to form a thin layer of BP, thin enough to transmit light with a high efficiency. Afterward, a small piece of the BP tape was cut out and attached onto a standard FC/PC fiber ferrule end surface with the aid of index matching gel. After connecting it with another FC/PC fiber ferrule with a standard flange adapter, the all-fiber BP based SA was finally ready. The entire preparation process is illustrated in Figure 4.1.

Figure 4.1: The preparation flow of BP-SA by using the mechanical exfoliation method. (a) The peeling BP crystal is place onto a sticky tape. (b) The crystal is thinned to the small flakes by repeatedly pressing the flakes onto a sticky tape. (c) The image of the BP tape after the thinning process. (d) Attaching the BP tape onto the ferrule.
The bandgap of a single layer of BP is relatively higher compared to the multi-layer. This leads to the use of a higher photon energy to excite the electron into the conduction band. The single layer needs a high energy for electron excitation due to the increase in its bandgap and has only 2.8% absorption of a single layer. However, multi-layer BP has a relatively narrow direct bandgap and that was considered a promising saturable absorber for the long wavelength range and has a higher modulation depth (F. Wang, 2017).

The composition of the multi-layer BP tape is examined by the energy dispersive spectroscopy (EDS) on the Field Emission Scanning Electron Microscopy (FESEM). Presence of the BP material on the scotch tape adhesive surface was confirmed by the presence of the high peak of phosphorus in the spectroscopy as shown in Figure 4.2. The inset in Figure 4.2 shows the FESEM image, which confirms the existence of uniform multi-layer phosphorus and the absence of > 1 µm aggregates or voids in the tape, which otherwise result in non-saturable scattering losses.

Raman spectroscopy was also performed on the prepared BP tape sample. Figure 4.3 shows the Raman spectrum, which is recorded by a spectrometer when a 514 nm beam of a Argon ion laser is radiated on the tape for 10 ms with an exposure power of 10 mW. As shown in the figure, the sample exhibits three distinct Raman peaks at 359 cm$^{-1}$, 434 cm$^{-1}$ and 460 cm$^{-1}$ (Sugai et al., 1985), corresponding to B$_{2g}$ and vibration modes of a layered BP. While B$_{2g}$ and modes correspond to the in-plane oscillation of phosphorus atoms in the BP layer, the mode corresponds to the out-of-plane vibration. The thickness for single-layer BP is about 0.8 nm (Andres et al., 2014), thus the prepared BP based SA is expected to consist of 250 layers.
Figure 4.2: EDF data from the FESEM image of the prepared BP tape, which confirms the presence of phosphorus. Inset: the FESEM image

Figure 4.3: Raman spectrum of the multi-layered BP tape

4.2.2 Nonlinear Saturable Absorption of BP-SA

The nonlinear optical response property for the multilayer BP on the scotch tape was also investigated to confirm its saturable absorption by applying an absorption technique. A self-constructed mode-locked fiber laser (1557 nm wavelength, 1.5 ps pulse width, 17.4
MHz repetition rate) was used as the input pulse source. The transmitted power and a reference power for normalization are recorded as a function of incident intensity on the tape by varying the input laser power. With increasing peak intensity, the material absorption decreases as shown in Figure 4.4, confirming the saturable absorption characteristic of the multi-layer BP. The experimental data for absorption are fitted according to a simple two-level SA model (Mu et al., 2015),

$$\alpha(I) = \frac{\alpha_s}{1 + I/I_{sat}} + \alpha_{ns}$$

(4.1)

where $\alpha(I)$ is the absorption, $\alpha_s$ is the modulation depth, $I$ is the input intensity, $I_{sat}$ is the saturation intensity, and $\alpha_{ns}$ is the non-saturable absorption. As shown in Figure 4.4, the modulation depth, non-saturable intensity, and saturation intensity are 7 %, 58 % and 0.0025 MW/cm$^2$, respectively. Taking into account its nonlinear optical response leading to absorption saturation at relatively low fluence, the mechanically exfoliated BP meets basic criteria of a passive SA for fiber lasers.

![Figure 4.4: Nonlinear saturable absorption profile of the multi-layer BP](image.png)
4.2.3 Experimental Setup for the Q-switched YDFL

The experimental setup of the proposed Q-switched YDFL is schematically shown in Figure 4.5. The setup is almost similar the proposed MoS$_2$ based Q-switched YDFL as described in Figure 3.3. The ring cavity consists of a 1.5 m long YDF, a wavelength division multiplexer (WDM), an isolator, a 3dB output coupler and the multi-layer BP nanomaterial based Q-switcher. The total cavity length is about 6 m. The YDF used has a core diameter of 4.0 µm, numerical aperture (NA) of 0.20 and cutoff wavelength of around 980 nm.

The doping level of ytterbium ions in the fiber is 1500 ppm. The YDF is pumped by a 980 nm pump laser diode through a 980/1030 nm WDM. The multilayer BP on the scotch tape is integrated into the cavity as a passive Q-switcher while a polarization insensitive optical isolator is incorporated to ensure unidirectional operation of the laser. The Q-switched pulse train is taken out via a 3dB output coupler which keeps half of the light oscillating in the ring cavity for both spectral and temporal diagnostics.

![Figure 4.5: Configuration of the proposed BP based Q-switched YDFL](image-url)
The output is simultaneously monitored using a 350-MHz oscilloscope together with a 2-GHz photodetector, a 7.8 GHz radio-frequency (RF) spectrum analyzer and an optical spectrum analyzer (OSA) with a spectral resolution of 0.02 nm. It is worth noting that the Q-switching performance such as output pulse energy of the laser is observed to be optimum with the use of the 3dB coupler.

4.2.4 Q-switching performance

The YDFL started to operate in the continuous-wave mode at the pump power of 40.5 mW, before transiting to the Q-switching operation at 55.1 mW. When the pump power was gradually increased up to 97.6 mW, a stable pulse train with an increasing repetition rate was observed. This is a typical feature of passive Q-switching. Figure 4.6 shows the spectral and temporal characteristics of the output Q-switching pulse train when the 980 nm pump power was fixed at 76.6 mW. The output spectrum is depicted in Figure 4.6 (a), which indicates that the Q-switched laser operates at 1069.4 nm with 3 dB spectral bandwidth of about 1.0 nm and signal to noise ratio of about 40 dB. Figure 4.6 (b) shows the typical oscilloscope trace of the Q-switched pulse trains. The pulse train has the period of 38.3 μs without noticeable timing jitter.

Figure 4.6 (c) reveals a typical pulse envelope, displaying an almost symmetrical Gaussian-like shape with a pulse duration of 12.2 μs. The RF output spectrum in Figure 4.6 (d) shows that the Q-switched pulses have a repetition rate of 26.1 kHz, which agrees with the pulse period of 38.3 μs measured in Figure 4.6 (b). It is also observed that the rf signal-to-noise ratio is about 23 dB with no spectral modulation, indicating that the passively Q-switching operation was stable.
Figure 4.6: Spectral and temporal characteristics of the output Q-switching pulse train at pump power of 76.6 mW (a) output spectrum (b) typical pulse train (c) typical single pulse envelop (d) RF spectrum
Figure 4.6, continued

Figure 4.7 shows the recorded pulse repetition rate and the pulse duration as a function of the pump power. The repetition rate of the Q-switching pulses linearly increases from
8.1 to 32.9 kHz as the pump power is increased from 55.1 to 82.0 mW. However, the repetition rate reduces as the pump power is further increased before the pulse train becomes unstable and disappears as the pump power is increased above 97.6 mW. This is due to the degradation of BP SA efficiency since the pump power increment increases the heat in the cavity.

Meanwhile the pulse duration significantly decreases from 17.9 to 10.8 μs as the pump power is increased from 55.1 to 97.6 mW. The pulse duration could be further narrowed by optimizing the parameters such as shortening the cavity length and improving the modulation depth of the BP Q-switcher. It is observed in Figure 4.8 that both average output power and pulse energy almost linearly increase with the input pump power up to the pump power of 97.6 mW. At 82 mW pump power, the pulse energy is changed due to the pulse width of the output laser that exhibits the increment trend at this pump power region. The maximum average output power and pulse energy are 10 mW and 328 nJ, respectively.

The experiment is also repeated for different locations of the BP tape sample. It found that a small deviation of tape or BP layer thickness does not change the optical properties of the BP SA and the Q-switched performance. We believe that the pulse energy produced by the laser could be further improved by optimizing the SA parameters of BP tape and the cavity design. The output pulse energy could be further improved if the cavity loss and SA loss can be further improved. The similar performance of Q-switching is also obtained in the erbium-doped fiber laser (EDFL) and the thulium-doped fiber laser (TDFL) setups, which operates at 1550 nm and 1920 nm, respectively. This shows that the proposed BP SA can be used as a wideband SA.
Figure 4.7: Repetition rate and pulse width of the proposed Q-switched YDFL against the pump power

Figure 4.8: Output power and pulse energy of the proposed Q-switched YDFL against the pump power
4.3 Mode-Locked YDFL with BP based SA

As previously discussed, ultra-short optical pulse sources have attracted widespread interests because of their useful applications such as micromachining, communication, and optical systems (Bao et al., 2009; Norihiko Nishizawa, 2014; N Nishizawa et al., 2008). A passive mode-locked method is implemented to generate pulse laser due to their nature being compact, low cost and easy to setup (Grelu et al., 2012; M. Zhang et al., 2009). Furthermore, active mode-locked method requires an additional switching electronics device in the laser cavity such as acousto-optic modulators (Ryu et al., 2007). Passively mode-locked fiber lasers are also widely explored in recent years by utilizing saturable absorption of optical materials. Various SAs have been demonstrated so far such as SESAMs (Gomes et al., 2004; J. Liu et al., 2012), carbon nanotubes (CNTs) (Solodyankin et al., 2008; J Sotor et al., 2015), and 2D materials including graphene (Huang et al., 2014; Z. Sun et al., 2010), TIs and transition metal dichalcogenides (TMDCs) (Dou et al., 2014; Sotor, Sobon, Grodecki, et al., 2014). TMDCs have a large bandgap that is more suitable for visible band (M. Ahmed, A. Latiff, H. Arof, & S. Harun, 2016; Q. H. Wang et al., 2012).

To date, another 2D nanomaterial, BP is also draw more interests as a promising material for ultra-short pulse applications (Park et al., 2015; Song et al., 2017; Jaroslaw Sotor et al., 2015; H. Yu et al., 2015). The reasons for this growing interest is because it has the most thermo-dynamically stable allotrope of phosphorus and a direct energy band structure in any case of its thickness, which can give excellent optical laser properties (Churchill et al., 2014; Lu et al., 2015). Compared to other 2D materials, BP has a direct band gap changing from 0.3 to 2.0 eV in BP layer. The advantage of bandgap is helpful for its optoelectronics applications and that make it more suitable for nonlinear short pulse modulation and can use as a saturable absorber to generate a mode-locking fiber laser (M. Ahmed, A. Latiff, H. Arof, & S. Harun, 2016; Han et al., 2017).
In this section, we have reported a mode-locked YDFL using a fabricated 2D material by the mechanically exfoliated technique of BP incorporate on end surface of a standard FC/PC fiber connector as SA. The mode-locked YDF laser operates at 1033.76 nm wavelength with 10 MHz repetition rate and 2.7 nJ pulse energy.

4.3.1 SA Preparation Process

In our experiment, the multi-layer BP SA was prepared by a mechanical exfoliation method as described in Figure 4.9. Mechanical exfoliation method in 2D materials has been generally employed for generation pulse fiber laser applications, such as graphene and TIs (Martinez et al., 2011; Sotor, Sobon, Macherzynski, et al., 2014), because of its reliability and simple in the fabrication process, which do not have any chemical procedures. First, thin flakes were moderately peeled off from a big block of commercially available BP crystal (purity of 99.995 %) using clear scotch tape. Then, the flakes were repeatedly pressed so that they adhered onto the scotch tape to form a thin layer of BP. Afterward, a fresh standard FC/PC fiber end surface was pushed down on the scotch tape so that the multilayer BP can be transferred onto it. With the aid of index matching gel, the ferrule with BP layers was connected to another fresh FC/PC fiber ferrule with a standard flange adapter. Then, the all-fiber BP based SA was finally ready.

![Figure 4.9: The fabrication process of mechanical exfoliation BP based SA](image-url)
Figure 4.10 shows the Raman spectrum, which is recorded of Argon ion laser is radiated on the tape for 10 ms with an exposure power of 10 mW. As shown in the figure, the sample exhibits three distinct Raman peaks at 360 cm\(^{-1}\), 438 cm\(^{-1}\) and 465 cm\(^{-1}\), corresponding to \(A_g^1\), \(B_{2g}\) and \(A_g^2\) vibration modes of a layered BP. While \(B_{2g}\) and \(A_g^2\) modes correspond to the in-plane oscillation of phosphorus atoms in BP layer, the \(A_g^1\) mode corresponds to the out-of-plane vibration.

![Raman spectrum of the BP based SA](image)

**Figure 4.10: Raman spectrum of the BP based SA**

The nonlinear optical response property for the BP SA was also experimentally investigated using an absorption technique in order to confirm its saturable absorption level. The nonlinear characterization was carried out using the same setup and approach as described in the previous section. The transmitted power and also a reference power for normalization are recorded as a function of incident intensity on the SA by varying the input laser power. With increasing peak intensity, the material absorption decreases as shown in Figure 4.11, confirming the saturable absorption characteristic of the multi-layer BP. The experimental data for absorption are fitted according to a simple two-level SA model as previously described in Equation (4.1). As shown in Figure 4.11, the
modulation depth, non-saturable intensity, and saturation intensity are 7.5 %, 57.5 % and 0.35 MW/cm², respectively. Taking into account its nonlinear optical response leading to absorption saturation at relatively low fluence, the mechanically exfoliated BP meets basic criteria of a passive SA for fiber lasers.

![Graph showing nonlinear absorption profile of BP SA](image)

**Figure 4.11: Nonlinear absorption profile of BP SA**

### 4.3.2 Experimental Setup for the Mode-locked YDFL

In this work, the performance of the BP SA is investigated for mode-locking application in YDFL cavity. Figure 4.12 shows the experimental setup, which mainly consists of YDF with length of about 1.5 m as a gain medium. The YDF (Yb1200-4/125) has a core and cladding diameters of 4 µm and 125 µm respectively, with a numerical aperture of 0.20, as well as the cut-off wavelength of 1010 nm and ion absorption of 280 dB/m at 920 nm. In order to ensure unidirectional operation, an isolator is spliced into the cavity. The 980 nm laser diode (LD) is coupled into the fiber ring resonator with a 980/1064 nm WDM. A 10 dB output coupler was used in the resonator to extract 10% of the output for analysis while allowing 90% of the light to oscillate in the laser cavity. A
polarization controller (PC) was incorporated inside the laser cavity between the SA device and isolator to adjust the polarization state of the oscillating light and optimizes the laser mode-locking performance. The output laser is analyzed by an OSA with a spectral resolution of 0.02 nm, an optical power meter and a 2 GHz photodetector together with a 350 MHz oscilloscope or 7.8 GHz RF spectrum analyzer. The laser cavity consists of 1.5 m YDF with a group velocity dispersion (GVD) of 24.22 ps\(^2\)/km and a single-mode fiber (SMF, HI 1060) with a total length of 19.5 m with GVD of 21.91 ps\(^2\) /km as the other fiber component. The net cavity dispersion was normal with a value estimated to be 0.46 ps\(^2\).

![Experiment configuration of mode-locked YDFL](image)

**Figure 4.12: Experiment configuration of mode-locked YDFL**

### 4.3.3 Performance of the Mode-locked Laser with BP SA

At first, the laser cavity was tested without incorporation of the SA device to confirm that there is no pulse obtained on the oscilloscope. Then the prepared BP SA is incorporated into the laser cavity as a mode-locker. As the pump power reaches 123.7 mW threshold, a continuous-wave laser was firstly obtained. A stable mode-locking pulse
was obtained as the pump power is further increased to 130.8 mW. The pulses train operating at repetition rate of 10 MHz was easily achieved by carefully controlling the rotation of the PC as the pump power is fixed within 130.8 mW to 200 mW. Figure 4.13 shows output spectrum of the mode-locked pulses at pump power of 200 mW. The spectrum shows a central wavelength of 1033.76 nm and 3dB spectral bandwidth of 0.48 nm with no Kelly sideband, which confirmed the operation of laser in normal dispersion. The narrow spectral bandwidth is due to the minimal nonlinearity and high net cavity dispersion. The Q-switching operation was also realized in the YDFL with the BP SA by increasing the cavity loss. It was achieved by replacing the 90/10 output coupler with a 3dB coupler. The Q-switched laser has a pump threshold of 55 mW, a pulse repetition rate that is tunable from 8 to 33 kHz, and the narrowest pulse width of around 10 μs.

Figure 4.14 shows the typical pulses train trace of the mode-locked YDFL measured by oscilloscope with pulse period of 96 ns at two different pump powers of 151.5 mW and 200 mW. It indicates there is no fluctuations in the pulse train. The achieved repetition rate pulse of 10 MHz is matched with the cavity length of 21 m. As shown in the same figure with the limitation of the oscilloscope resolution, the achieved pulse width of 49 ns is much broader than the actual pulse width. As an alternative, the pulse width can be measured by using calculation based on the time bandwidth product (TBP) formula. A mathematically estimated a minimum possible pulse width of approximately 3.27 ps with TBP of 0.441 for Gaussian pulse profile and 3dB spectral bandwidth of 0.48 nm. The RF spectrum for the proposed YDFL is shown in Figure 4.15, which has confirmed that the mode-locked has a fundamental frequency of 10 MHz with a signal-to-noise ratio (SNR) up to 39 dB. By removing out the BP SA from the ring cavity, the mode-locking operation could not be achieved, no matter how we adjust the PC and pump power. This verified that the mode-locking operation is purely caused by the BP SA. These results show that
black phosphorus and other similar 2D materials may have a good potential for application in ultrafast photonics.

Figure 4.13: Optical spectrum of the proposed mode-locked YDFL at pump power of 200 mW

Figure 4.14: Typical pulse train of mode-locking YDFL at two different pump powers
Figure 4.15: RF spectrum of the mode-locked YDFL

Figure 4.16 shows the graphs of an average output, pulse energy and peak power as a function of input pump power for the mode-locked YDFL. As seen, the average output power, pulse energy, peak power rise linearly with the pump power. The average output power is increased from 4.7 to 27 mW as the pump power is increased from 130.8 to 200 mW as shown in Figure 4.16 (a). The pulse energy is measured based on the output power and repetition rate as shown in the Figure 4.16 (b). The maximum pulse energy is 2.7 nJ at the maximum pump power of 200 mW. Figure 4.16 (c) shows the peak power against the pump power, which the maximum peak power is 0.83 kW at 200 mW pump power. In order to examine the long term stability of the mode-locked laser, we continuously monitor the output signal by an optical spectrum analyzer and an oscilloscope for 48 hours. It is observed that central wavelength and 3 dB bandwidth of the spectrum are almost unchanged while the repetition rate stayed constant throughout the experiment. This observation proves the stability of BP as a SA in mode-locked fiber laser.

BP has a direct transition for all thicknesses, which shows a significant benefit for optoelectronic applications, especially at the long-wavelength range. As it can offer a
broad tuning range for a bandgap with number of layers, and can bridge nearly the entire range between the zero bandgap of graphene and the large bandgap of TMDCs. Furthermore, the preparation of the BP is also simple and thus it is preferable compared to TIs, which limit themselves by the complex fabrication processes. However, the BP materials are very hydrophilic and thus the SA is easily damaged with the exposure to oxygen and water molecules.

Nevertheless, the BP SA remain undamaged as we tuned pump power up to 200 mW for more than 2 days. By keeping in an air tight container, less expose to oxygen or water molecules helps to protect BP SA from damage easily. The experimental results verify the mode locking ability of the newly developed BP based SA. This shows that the BP could be used to develop promising optoelectronic devices with high power tolerance, offering inroads for more practical applications, such as large energy laser mode-locking, nonlinear optical modulation and signal processing etc.

![Graph of output power versus pump power](image-url)

**Figure 4.16**: Variation in (a) output power, (b) pulse energy, and (c) peak power at different pump power
Figure 4.16, continued

(b) Pulse energy (nJ) vs. Pump power (mW)

(c) Peak power (kW) vs. Pump power (mW)
4.4 Summary

Multi-layer BP have been successfully demonstrated as an effective SA for both passive Q-switching and mode-locking applications in YDFL cavity. The BP SA was prepared by mechanical exfoliation method and sandwiched between two fiber ferrules via a fiber adapter to form a fiber-compatible SA. Firstly, a stable Q-switched pulses at 1069.4 nm were successfully obtained using the SA tape with BP layers. The laser registered a pump threshold of 55.1 mW and the minimum pulse duration was 10.8 μs. The pulse repetition rate could be varied over a wide range of frequencies, from 8.2 to 32.9 kHz, by adjusting the pump power. The highest pulse energy of 328 nJ was obtained at pump power of 97.6 mW. Secondly, passively mode-locked YDFL was demonstrated using an improved multi-layer BP based SA, which was obtained without the sticky tape. The laser cavity was designed with normal net cavity dispersion of 0.46 ps² while inserting additional SMF and PC in the cavity. The proposed mode-locked YDF laser operated at 1033.76 nm wavelength with a repetition rate of 10 MHz. The highest pulse energy and peak power was measured to be around 2.7 nJ and 0.83 kW respectively at pump power of 200 mW. The fabrication of BP SA is easy, cheap and suitable device to generate mode-locked laser in 1 μm wavelength region. These experimental results suggest that a multi-layer BP is a promising material for pulsed laser applications.
CHAPTER 5: TOPOLOGICAL INSULATOR AS SATURABLE ABSORBER

5.1 Introduction

Topological insulators (TIs) have attracted attention of researches due to the high nonlinear susceptibility and large modulation depth with efficient saturable absorption property. They are excellent semiconductors with high surface electron mobility. The mostly used TIs are Bismuth (III) Selenide ($\text{Bi}_2\text{S}_3$), Bismuth (III) Telluride ($\text{Bi}_2\text{Te}_3$) and Antimony (III) Telluride ($\text{Sb}_2\text{Te}_3$), which have a band gap of 0.35, 0.21, and 0.30 eV, respectively and the physical properties of these three TIs are similar because they are composed of two elements (W. Tian et al., 2017). These materials have a thickness of nano-to micro scale (Junsu Lee, Koo, Jhon, et al., 2014) and they show a linear dispersion relation at low pulse energy and nonlinear dispersion relation at the excitation of high energy pulse and thus suitable for saturable absorber (SA) application. More recently, passive SAs made of topological insulators (TIs) have been shown to be capable of generating Q-switched and mode-locked lasers (Y. Chen et al., 2014; H. Liu, X.-W. Zheng, et al., 2014). Since TIs have a large modulation depth with an efficient saturable absorption property, they are suitable for making SAs. For instance, $\text{Bi}_2\text{Te}_3$ has the modulation depth and saturation intensity of about 22% and 57 MW/cm² respectively (Y. Chen et al., 2014).

In this chapter, passively Q-switched Ytterbium-doped fiber laser (YDFL) are demonstrated using a few-layers $\text{Bi}_2\text{Se}_3$, $\text{Bi}_2\text{Te}_3$ and antimony telluride ($\text{Sb}_2\text{Te}_3$) based SAs while mode-locked YDFL is demonstrated using $\text{Sb}_2\text{Te}_3$ material to exploit the wideband saturable-absorption characteristic of the TIs. For this purpose, we prepare free-standing $\text{Bi}_2\text{Se}_3$-, $\text{Bi}_2\text{Te}_3$- and $\text{Sb}_2\text{Te}_3$-polymer composite SA films by implanting the powder of these materials into a polyvinyl alcohol (PVA) host. These films are used in a fully fiber-integrated laser cavity for generating pulses train.
5.2 Q-switching Performance of YDFL with Few-layer Bi$_2$Se$_3$ and Bi$_2$Te$_3$ based SAs

Recently, passive SAs made of topological insulators (TIs) have been shown to be capable of generating Q-switching pulses train (Y. Chen et al., 2014; H. Liu, X.-W. Zheng, et al., 2014). For instance, Q-switched Erbium-doped fiber lasers (EDFLs) were demonstrated using Bismuth (III) Selenide (Bi$_2$Se$_3$) and Bi$_2$Te$_3$ SAs (Ahmad et al., 2015; Y. Chen et al., 2014). Chen et al. (2014) demonstrated Bi$_2$Se$_3$ based Q-switched EDFL with repetition rates range from 4.5 to 12.9 kHz and the shortest pulse width of 13.4 µs (Y. Chen et al., 2014). Luo et al. used Bi$_2$Se$_3$ fabricated by the liquid-phase exfoliation method as SA in YDFL cavity to generate Q-switching pulses with repetition rate ranging from 8.3 to 29.1 kHz, shortest pulse width of 1.95 µs and highest pulse energy of 17.9 nJ (Z. Luo et al., 2013). The SA used has modulation depth of 3.8% and saturable intensity of 53 MW/cm$^2$.

In this section, two different passively Q-switched YDFLs are demonstrated using Bi$_2$Se$_3$ and Bi$_2$Te$_3$ nanoparticles based SAs to exploit the wideband saturable-absorption characteristic of the TIs. Compared with the previous YDFL (Z. Luo et al., 2013), higher repetition rate is obtained in both fiber lasers.

5.2.1 Preparation and Characterization of Bi$_2$Se$_3$ and Bi$_2$Te$_3$ SAs

The commercially available TI powders of Bi$_2$Se$_3$ and Bi$_2$Te$_3$ with molecular weight of 654.84 g/mol and 800.76 g/mol respectively, are used to prepare the SA in this experiment. The fabrication procedures for both TI films are described in Figure 5.1. To prepare the host polymer, 1 g of polyvinyl Alcohol (PVA) (Sigma Aldrich) is dissolved in 120 ml de-ionized (DI) water with the aid of a magnetic stirrer at room temperature. Next, 14 mg of Bi$_2$Se$_3$ powder and 14 mg of Bi$_2$Te$_3$ powder are mixed with 3 ml of the PVA solution separately. They are thoroughly mixed using a magnetic stirrer for three
hours. Then the Bi$_2$Se$_3$-PVA and Bi$_2$Te$_3$-PVA solutions were placed in ultrasonic bath for 10 minutes to make sure the Bi$_2$Se$_3$ and Bi$_2$Te$_3$ powder fully binds with the PVA. After that, both TI suspensions are carefully poured onto petri dishes to avoid trapping any air bubble and is left to dry at room temperature for 48 hours to form Bi$_2$Se$_3$-PVA and Bi$_2$Te$_3$-PVA composite films. The thickness of Bi$_2$Se$_3$ and Bi$_2$Te$_3$ films are about 55 and 68 µm, respectively.

Figure 5.2 (a) and (b) shows the field emission scanning electron microscopy (FESEM) images of Bi$_2$Se$_3$ and Bi$_2$Te$_3$ film, respectively. As shown in Figure 5.2 (a), the Bi$_2$Se$_3$ film has a high dense of micro-rods and micro-grains, which can be clearly viewed on the substrate surface and distributed randomly on the substrate surface. These micro-rods and micro-grains are in irregular shapes with an average size of 0.3 to 1.66 µm. As shown in Figure 5.2 (b), the Bi$_2$Te$_3$ film has also a high dense of acicular grains formation like micro-networks, which are in irregular shapes in a size between 0.5 to 1.9 µm.

Figure 5.1: The fabrication process of Bi$_2$Se$_3$ and Bi$_2$Te$_3$ thin film
Figure 5.2: FESEM images of (a) the Bi$_2$Se$_3$ and (b) Bi$_2$Te$_3$ composite film. Insert of each image represent a high magnification of the respective image.

Figure 5.3 illustrates the absorbance spectrum of the prepared PVA only, Bi$_2$Se$_3$-PVA and Bi$_2$Te$_3$-PVA films in the range of 200 nm to 1100 nm. The figure attests a constant absorbance for both Bi$_2$Se$_3$-PVA and Bi$_2$Te$_3$-PVA film which indicates that they possess a broadband resonance wavelength like graphene. It is shown that the incorporation of Bi$_2$Se$_3$ and Bi$_2$Te$_3$ nanoparticles into the PVA increase the loss of the film. The Bi$_2$Te$_3$ film is slightly thicker and thus it has a higher loss compared to the Bi$_2$Se$_3$ film. However,
at around 1100 nm, both Bi₂Se₃ and Bi₂Te₃ films suffer an almost identical absorption loss. It also shows that PVA polymer is an excellent host material for fabricating high performance TI SA and not affected by this host polymer. Figure 5.4 shows the Raman spectra of the fabricated films. For Bi₂Se₃-PVA film, the three distinct peaks observed at ~67 cm⁻¹, ~126 cm⁻¹, and ~170 cm⁻¹ can be assigned to the A¹₁g, E₂𝑔 and A²₂𝑔 vibrational modes, respectively. It has also shown that the Raman spectrum of the developed Bi₂Te₃-PVA film has three peaks at ~67 cm⁻¹(A¹₁g), ~96 (E₂𝑔) and 118(A²₁g) with a lower power. The lower peak power is most probably due to the lower concentration of the Bismuth (III) Telluride powder in PVA and the effect of the film thickness (Russo et al., 2008). In this experiment, the same ratio of Bi₂Se₃ and Bi₂Te₃ of 14 mg was used in 3 ml PVA, but Bi₂Te₃ powder has a higher molecular weight (800.76 g/mol) compared to Bi₂Se₃ (654.84 g/mol). To improve the Raman resonant in the Bi₂Te₃-PVA film, the weight of Bi₂Te₃ powder could be increased. The simple fabrication method for both Bi₂Se₃ and Bi₂Te₃ PVA films in room temperature is very attractive for scalable production.

Figure 5.3: Linear transmission of the free standing Bi₂Se₃-PVA and Bi₂Te₃-PVA films
The nonlinear optical response property for the Bi$_2$Se$_3$ and Bi$_2$Te$_3$ were also investigated to confirm their saturable absorption by applying the similar technique as described in the previous chapter. The pulse input source used a mode-locked fiber laser, which has femtosecond output pulse with a 17 MHz repetition rate and a 900 fs pulse duration, which the output power is approximately 5 mW. Figure 5.5(a) and (b) show the transmission nonlinear characteristics of the Bi$_2$Se$_3$ and Bi$_2$Te$_3$ SAs, respectively, which were fitted using the following simple saturable absorption model of

$$T(I) = 1 - \alpha_0 \exp \left( -I/I_{sat} \right) - \alpha_{ns}$$  \hspace{1cm} (5.1)

Here, $T(I)$, $\alpha_0$, $I$, $I_{sat}$ and $\alpha_{ns}$ stand for the transmission, modulation depth, input intensity, saturation intensity, and non-saturable absorption, respectively (Z. Tian et al., 2015). For Bi$_2$Se$_3$ SA, the modulation depth, non-saturable absorption and saturation intensity were measured to be approximately 15.9 %, 77 % and 50 MW/cm$^2$, respectively as shown in Figure 5.5 (a). As well as in Bi$_2$Te$_3$ SA shown in Figure 5.5 (b), the

Figure 5.4: Raman spectrum for the free standing Bi$_2$Se$_3$-PVA and Bi$_2$Te$_3$-PVA films

![Raman spectrum](image-url)
modulation depth, non-saturable absorption and saturation intensity were obtained to be approximately 30 %, 55 % and 40 MW/cm², respectively. In this case, the modulation depth of Bi₂Te₃ sample is higher, however the saturation intensity is smaller than in the Bi₂Se₃ sample.

![Figure 5.5: Characteristics of nonlinear absorption measurement for (a) Bi₂Se₃-PVA film, and (b) Bi₂Te₃-PVA film](image-url)
5.2.2 Experimental Arrangement

The schematic experimental setup of the proposed Q-switched YDFL with TI based SA is depicted in Figure 5.6. The ring laser consists of a 1.5 m long YDF as the active medium, a 980/1064 nm wavelength division multiplexer (WDM), an optical isolator, a TI: Bi$_2$Se$_3$ or TI: Bi$_2$Te$_3$ SA and a 10 dB output coupler. The YDF used has Ytterbium ions concentration of 1500 ppm, a core diameter of 4 µm, numerical aperture of 0.20 and cut-off wavelength of 950 nm. It is pumped by a 980 nm laser diode through the WDM. The other end of the YDF is spliced to the 10 dB output coupler, which allows 10% of the output to be extracted for analysis. The remainder is channeled to the SA device and kept in the cavity to oscillate for laser generation. The SA device is fabricated by sandwiching a ~ 1 mm×1 mm piece of the Bi$_2$Se$_3$ or Bi$_2$Te$_3$ composite film between two fiber ferrules using a fiber adaptor. Index matching gel is applied at the connection to minimize parasitic reflections. The SA is connected to the isolator, which prevents backward reflection and ensures unidirectional operation. The other port of the isolator is spliced to the 1064 nm port of the WDM to link the ring cavity. The output laser is analysed by an optical spectrum analyzer (OSA) with a spectral resolution of 0.02 nm, an optical power meter and a 2 GHz photodetector together with a 350 MHz oscilloscope or 7.8 GHz radio-frequency (RF) spectrum analyzer. The total cavity length is around 11 m.

Figure 5.6: Experimental setup for the Q-switched YDFL with TI: Bi$_2$Se$_3$ or TI: Bi$_2$Te$_3$ based SA
5.2.3 The Performance of the Q-switched Lasers

The performance of the YDFL cavity was investigated by varying the power of the 980 nm pump. The continuous wave (CW) laser started as the power hit the threshold of 43.7 mW. Further, a self-starting and stable Q-switched operation was obtained when the pump power reached the threshold of 67.6 mW and 72.8 mW for Bi$_2$Se$_3$ and Bi$_2$Te$_3$, respectively. Figure 5.7 illustrates the output optical spectra of the Q-switched YDFL constructed with Bi$_2$Se$_3$ and Bi$_2$Te$_3$ based SA at 72.8 mW pump power. As shown, the output optical spectra of Bi$_2$Se$_3$ and Bi$_2$Te$_3$ based Q-switched YDFLs center at wavelengths of 1057.4, and 1044.5 nm with 3dB bandwidth of 1.32 and 1 nm, and peak output power of -30, and -27.46 dBm, respectively. The CW YDFL output spectrum, which was obtained by removing the SA is also shown for comparison. Without the SA, the YDFL operates at the center wavelength of 1062 nm. The operating wavelength shifts to a shorter wavelength with the employment of SA due to the film insertion loss. The Q-switched lasers operate at a shorter wavelength, which has a higher gain to compensate for the loss.

This result also indicates that the Bi$_2$Te$_3$ film has a slightly higher loss than Bi$_2$Se$_3$. Spectral broadening is also observed in the Q-switched lasers due to self-phase modulation (SPM) effect in the ring cavity. The same ratio of Bi$_2$Se$_3$ and Bi$_2$Te$_3$ of 14 mg in 5 ml PVA has used, however Bi$_2$Te$_3$ powder is with higher molecular weight (800.76 g/mol) compared to Bi$_2$Se$_3$ (654.84 g/mol). The Bi$_2$Se$_3$ powder has higher degree of dispersion in PVA compare to Bi$_2$Te$_3$ powder and this will affect the performance of the developed film as saturale absorber. Therefore, the bandwidth of output laser of Bi$_2$Se$_3$ is much wider than Bi$_2$Te$_3$ as shown in Figure 5.7.
Figure 5.7: Optical spectra of the CW and Q-switched YDFLs at pump power of 72.8 mW

The comparison performance of the Q-switched laser with Bi$_2$Se$_3$ and Bi$_2$Te$_3$ based SA is summarized in Figure 5.8. A stable Q-switching pulses train is generated by increasing the pump power above 67.6 mW until 88.3 mW for Bi$_2$Se$_3$ SA, while from 72.8 mW until 98.4 mW for Bi$_2$Te$_3$ SA. Figure 5.8 (a) displays the typical oscilloscope trace at the threshold pump power of both SAs. For Bi$_2$Se$_3$ SA, the typical oscilloscope shows at 67.6 mW and threshold pump power, which shows a pulse repetition rate of 48.83 kHz. The pulse train has an average output power of 0.65 mW which matches to the pulse energy of 13.3 nJ. With Bi$_2$Te$_3$ SA, the typical oscilloscope trace is shown at the threshold pump power of 72.8 mW, which has a repetition rate of 43.25 kHz and pulse width of 5.65 µs.

Figure 5.8 (b)-(c) compares the pulse train and the envelop of the single pulse at maximum pump power of Q-switching pulses obtained by the Bi$_2$Se$_3$ and Bi$_2$Te$_3$ SA at pump power of 88.3 mW, and 98.4 mW respectively. The repetition rate between two pulses are measured to be around 102 kHz and 93.63 kHz, whereas the maximum Q-
switching pulse envelope with a period of 3.48 µs and 2.01 µs as measured from the full-width half maximum (FWHM). There is no amplitude fluctuation for the pulse within the tunable range of the repetition rate for both SAs.

Figure 5.8 (d) shows the measured pulse repetition rate and pulse width against the pump power for both Q-switched lasers. It is shown that the pulse repetition rate linearly increases, while the pulse width linearly decreases with the pump power. This matches well with the passive Q-switched theory based on saturable absorber. The pulse repetition rate of Bi$_2$Se$_3$ SA based Q-switched YDFL linearly increases from 48.83 to 102 kHz while the pulse width decreases from 8.9 to 3.48 µs as the pump power rises from 67.6 to 88.3 mW. The Q-switching pulse disappears when the pump power is increased above 88.3 mW. Similarly, the pulse width of the Bi$_2$Te$_3$ based Q-switched YDFL decreases from 5.65 to 2.01 µs with the increase in pump power from 72.8 to 98.4 mW. The associated repetition rate increases from 43.25 to 93.63 kHz within the same range. This is typical of a Q-switched fiber laser due to the gain compression effect in the laser cavity. Compared to the Bi$_2$Se$_3$ laser, the Bi$_2$Te$_3$ Q-switched laser shows a narrower tuning range for both repetition rate and pulse width.

The pulse width YDFL of the Bi$_2$Se$_3$ is expected to drop further if the pump power can be increased more, as long as the damage threshold of the SA is not exceeded. The Q-switched threshold is also lower for the Bi$_2$Te$_3$ laser due to the thicker film layer, which allows more absorption. It is observed that the Q-switching pulse disappears when the pump power is increased above 88.3 mW and 98.4 mW for Bi$_2$Se$_3$ and Bi$_2$Te$_3$ SA, respectively. We observed pulses in certain range of the pump power, and at high pumping the pulse disappeared, due to the SA is continuously in the saturated state because of the high intensity of light, and there is no time for de-excitation. However, the
two Q-switching lasers are obtained again by reducing the pump power within the range of operation.

Figure 5.8 (e) shows the measured average output power and the calculated pulse energy of both Q-switched YDFLs as a function of pump power. As seen in the figure, the average output power further increases with the pump power, while the pulse energy fluctuates within the pump power for both Q-switching lasers. For Bi$_2$Se$_3$ based YDFL, the output power increases from 0.65 to 1.27 mW within the pump power range from 67.6 to 88.3 mW.

The maximum output power of 1.27 mW is obtained at the maximum pump power of 88.3 mW, while the maximum pulse energy of 16.6 nJ is recorded at 72.8 mW. In comparison, the average output power of the Bi$_2$Te$_3$ based YDFL linearly increases from 1 to 2.36 mW as the pump power grows from 72.8 to 98.4 mW. As the pump power increases, the non-radiative decay rate of the Yb$^{3+}$ ions also increases. This generates heat in the laser cavity system, which it’s absorbed by the SA. This will energize some electrons in the SA’s valance band to move up to the conduction band from the strong internal thermal motion. As fewer electrons in the valence band are available for photon absorption, the initial transmittance of the SA becomes greater as the pump power or the levels of absorber heat increases.

Thus, the SA’s efficiency as SA is greatly reduced, meaning that the pulse width begins to increase again after the saturation of SA. The maximum pulse energy of 29.95 nJ is obtained at 83.2 mW pump power. After increasing the pump power above 98.4 mW, the SA was working in a good condition, however no indication of mode-locked pulse. Thus, the value of damage threshold cannot be defined due to the limited pump power of 200 mW.
Figure 5.8: The performance of Q-switched YDFL with Bi$_2$Se$_3$ and Bi$_2$Te$_3$ based SA (a) pulse train at threshold pump power of 67.6 mW and 72.8 mW, respectively (b)-(c) Pulse train and single envelope pulse at maximum pump power of 88.3 mW, and 98.4 mW, respectively (d) Pulse width and repetition rate characteristics at various pump power. (e) The average output power and pulse energy characteristics at various pump power
Figure 5.8, continued

(c) Pulse width (μs)

Repetition rate (Bi₂Se₃)

Repetition rate (Bi₂Te₃)

Pulse width (Bi₂Se₃)

Pulse width (Bi₂Te₃)

(d) Repetition rate (Bi₂Se₃) vs Pump power (mW)

Repetition rate (Bi₂Te₃)

Pulse Width (Bi₂Se₃)

Pulse Width (Bi₂Te₃)
Figure 5.9 depicts the radio frequency (RF) spectra of the Q-switched YDFLs configured with Bi\textsubscript{2}Se\textsubscript{3} and Bi\textsubscript{2}Te\textsubscript{3} SA, which were achieved with resolution bandwidth of 3 kHz at the maximum pump powers of 88.3 and 98.4 mW, respectively. Both spectra were taken using an RF spectrum analyzer via a 1.2 GHz photodetector. The maximum repetition rate of 102 kHz and 93.63 kHz are obtained with the Bi\textsubscript{2}Se\textsubscript{3} and Bi\textsubscript{2}Te\textsubscript{3} SA, respectively. The corresponding signal to noise ratios (SNR) are 55 and 53 dB respectively. The high SNR values indicate the stability of both Q-switched lasers. The results show that both Bi\textsubscript{2}Se\textsubscript{3} and Bi\textsubscript{2}Te\textsubscript{3} have a great potential for use in generating Q-switching pulses. The fabrication of both SAs is simple and cheap while the output of both Q-switched lasers is highly stable at room temperature. The pulse width could be further narrowed by employing a shorter laser cavity and improving the modulation depth of the SA. It is found that the Bi\textsubscript{2}Te\textsubscript{3} SA produces a smaller minimum pulse width while
the Bi$_2$Se$_3$ SA produces a higher maximum repetition rate. In addition, the highest pulse energy of 29.95 nJ is obtained with the Bi$_2$Te$_3$ SA.

![Figure 5.9: RF spectra of the Q-switched YDFLs configured with Bi$_2$Se$_3$ and Bi$_2$Te$_3$](image)

5.3 Q-Switched and Mode-locked YDFLs with Antimony Telluride TI based SA

In the previous section, both Bi$_2$Se$_3$ and Bi$_2$Te$_3$ SAs show good Q-switching performance in the 1µm wavelength region. However, the mode-locking operation cannot be achieved by both SAs. This is most probably due to the modulation depth, dispersion and nonlinear properties of the fabricated SAs, which were not suitable for mode-locking operation. However, Sotor et al. demonstrated antimony telluride (Sb$_2$Te$_3$) as SA incorporated on a side-polished erbium-doped fiber to generate a passive mode-locking pulse with pulse duration of 128 fs and repetition rate of 22.32 MHz. Its modulation depth and non-saturation loss were 6% and 43%, respectively (Sotor, Sobon, & Abramski, 2014). In another work, Kowalczyk et al. studied the mode-locked YDFL using Sb$_2$Te$_3$ SA, which was fabricated by a pulsed magnetron sputtering technique. The YDFL with 2 mm long Sb$_2$Te$_3$ SA obtained at pulse width of 5.9 ps (Kowalczyk, Bogusławski, et al., 2016). In this section, the generation of Q-switched and mode-locked
YDFLs are demonstrated using a simpler technique based on the Sb$_2$Te$_3$ material deposited on the end-facet of a fiber ferrule as an SA device. By incorporating the Sb$_2$Te$_3$ SA into the YDFL cavity, Q-switched and mode-locked operation could be achieved.

### 5.3.1 Preparation and Characterization of Sb$_2$Te$_3$ film SA

In this experiment, the molecular weight of the Sb$_3$Te$_2$ is about 626.32 g/mol. In order to prepare the PVA host polymer, 1 g of PVA (Sigma-Aldrich) is dissolved in 120 ml deionized water with the aid of a magnetic stirrer at room temperature as shown in Figure 5.10. The SA was prepared by mixing 5 mg powder [Sigma Aldrich, -325 Mesh, 99.6% trace metal basis] into 5 ml of the PVA solution. Next, the mixture was stirred for 3 hours using magnetic stirrer and followed by ultra-sonication of an hour to prevent any aggregation. Then, the mixture of suspensions is carefully poured onto the petri dish to avoid trapping any air bubbles and then dry in the room temperature for 3 days to form Sb$_3$Te$_2$-PVA composite film. The thickness of Sb$_3$Te$_2$ is about 34 µm. The advantages of this process are to clean growth environment, low growth temperature, good crystal safety, regular thickness, and easy to dope (W. Tian et al., 2017).

![Diagram of fabrication process of Sb$_2$Te$_3$ PVA film](image)

**Figure 5.10: The fabrication process of Sb$_2$Te$_3$ PVA film**
Figure 5.11 (a) shows the FESEM image of a uniform polymeric thin film of Sb$_2$Te$_3$. Several bumps is observed on the surface, as seen in an insert figure, in like-spherical shape with a size between 0.2 and 1.33 µm. These formed bumps may have resulted after drying the thin film at room temperature. Figure 5.11 (b) shows the absorbance spectrum of the prepared Sb$_2$Te$_3$-PVA and PVA only in range from 200 to 1000 nm. It also shows that PVA polymer is an excellent host material for fabricating high performance Sb$_2$Te$_3$ SA and not affected by this host polymer. Figure 5.11 (c) shows the Raman spectrum of Sb$_2$Te$_3$-PVA film.

It was obtained using Renishaw InVia Raman microscope with a charge coupled device detector and argon-ion laser with a diffraction grating of 1200 lines/mm. the excitation wavelength and laser power were used at 514 nm and 10 mW, respectively. Two modes were shown at 2947.48 and 1433.18 cm$^{-1}$ which are attributed to valence C-H vibration and shear modes for PVA polymer, respectively (C. Wang et al., 2013). An inset figure shows four modes of Sb$_2$Te$_3$ at 49.52, 63.17, 106.72 and 171.68 cm$^{-1}$, corresponding to E$_g^1$ (TO), A$_{1g}^1$ (LO), E$_g^2$ (TO) and A$_{2g}^1$ (LO), respectively (Sotor, Sobon, Grodecki, et al., 2014) and a peak at 133.84 cm$^{-1}$, which is belong to Te-Te bonds (packet structure) (Bogusławski et al., 2016).

By applying dual-detector measurement technique, the nonlinear optical response property for the Sb$_2$Te$_3$ thin film was obtained as depicted in Figure 5.11 (d). It shows the recorded data of transmission at various input intensities for the Sb$_2$Te$_3$-PVA SA, which were fitted by using Equation 5.1. As seen in the figure, the modulation depth, non-saturable absorption, and saturation intensity were measured to be 8.5%, 70%, and 36 MW/cm$^2$, respectively.
Figure 5.11: The characteristics of the fabricated Sb$_2$Te$_3$ PVA based SA (a) FESEM images of the Sb$_2$Te$_3$–PVA composite thin film. Inset shows a high magnification of the image, (b) Linear transmission of the Sb$_2$Te$_3$–PVA as compared with PVA only free standing, (c) Raman spectrum, and (d) Nonlinear transmission measurement.
Figure 5.11, continued
5.3.2 Experimental Setup

The pulses generation in a YDFL ring cavity is investigated by exploiting the absorption property of Sb$_2$Te$_3$ SA. Figure 5.12 shows the experimental setup of the proposed Sb$_2$Te$_3$ SA based Q-switched YDFL. The laser cavity employs a 1.5 meter-long YDF, which was pumped by a 980 nm pump through a 980/1064 nm WDM as the gain medium. The YDF used is same with the previous work as described in the previous section. An isolator was integrated into the optical ring cavity to ensure unidirectional operation. A 3 dB optical coupler was employed to tap out the laser emission while keeping half of the power inside the cavity to oscillate for laser generation. The Sb$_2$Te$_3$ TI SA device was fabricated by sandwiching a ~1 mm×1 mm piece between two fiber ferrules using a fiber adaptor and index matching gel was applied at the connection to minimize parasitic reflections.

![Experimental Setup Diagram](image)

**Figure 5.12:** The schematic diagram of Sb$_2$Te$_3$-PVA SA based Q-switched YDFL. The configuration was converted into a mode-locked laser by employing 90/10 output coupler instead of 50/50 coupler.
The output laser was analyzed by an OSA with a spectral resolution of 0.02 nm, an optical power meter and a 2 GHz photodetector together with a 350 MHz oscilloscope or 7.8 GHz RF spectrum analyzer. The laser cavity was then converted into a mode-locked laser by replacing the 3dB coupler with 90/10 coupler. The mode-locking pulses train was tapped out from 10% port while allowing 90% of the light to oscillate in the cavity. The mode-locking operation was achieved since the cavity loss was significantly reduced by the proposed arrangement.

5.3.3 Q-switching Laser Performances

In Q-switched laser cavity, a 3dB coupler was employed and the output pulses were channeled out through one of the output coupler port for analyzing. A continuous wave (CW) operation was observed when the pump power reached to 61.5 mW. A self-started Q-switching operation was achieved at 75.4 mW pump power and the Q-switching operation was maintained up to the pump power of 96.2 mW. Unstable Q-switched state was observed when the pump power was increased higher than 96.2 mW. This is attributed to the Sb$_2$Te$_3$ SA, which was over-saturated at high incident intensity. However, there is no damage in SA under high pump power and the Q-switched state is always obtained as the pump power is reduced back within a range between 75.4 and 96.2 mW.

Figure 5.13 shows the evolution of Q-switching pulses train with increasing in pump power. As seen, the pulse repetition rate is 24.4, 28.38, 32.5 and 51.44 kHz at pump power of 75.4, 82.3, 85.8 and 92.7 mW respectively. It indicates the feature of passive Q-switching state where the repetition rate is progressively increased with pump power increment.
Figure 5.13: Typical pulse train of the Q-switched laser at different pump powers

Figure 5.14 summarizes the typical performance of Q-switching pulse state emitted from YDFL using Sb$_2$Te$_3$ SA at maximum pump power of 96.2 mW. Figure 5.14 (a) displays the typical Q-switching oscilloscope trace, which has a repetition rate of 55 kHz and the pulse period (peak to peak spacing) of around 18.2 µs. A single pulse profile has a full-width half maximum (FWHM) of 6.6 µs at the maximum pump power as shown in Figure 5.14 (b). There is no amplitude fluctuation for the pulse within the tunable range of the repetition rate and no peak intensity modulation on pulse train, which indicates the high stability of the Q-switching state. Figure 5.14 (c) shows the optical spectrum of Q-switched YDFL, which operates at wavelength of 1070.2 nm with peak output power of -4.5 dBm. In order to investigate the stability of Q-switching pulses train in the proposed Sb$_2$Te$_3$ SA based YDFL, RF spectrum was obtained at this pump power using an RF spectrum analyzer via a 1.2 GHz photodetector as shown in Figure 5.14 (d). The signal to noise ratio (SNR) obtained was about 60 dB, which indicates the excellent stability.
Figure 5.14: Typical performance of Q-switching pulse emitted from our YDF laser using Sb$_2$Te$_3$ SA at pump power of 96.2 mW: (a) Oscilloscope pulse train, (b) Single pulse profile, (c) optical spectrum, (d) RF spectrum

Figure 5.15 (a) shows the measurement of repetition rate and pulse width at various pump power for the proposed Q-switched YDFL. It can be seen that the repetition rate linearly increases, while the pulse width is decreased with increasing in pump power. As the pump power increases from 75.4 to 96.2 mW, the pulse repetition rate increases from 24.4 to 55 kHz, while the pulse width decreases rapidly from 9.8 to 6.6 µs. This is most probably due to the fast accumulation of electrons in the upper energy level. Yet, in the high pump power regime, according to the over-saturation, the speed of accumulation slows down, and leading to less significant change of pulse widths. This is a typical characteristic for a Q-switched laser, which mainly due to the gain compression effect in the laser cavity. It is observed that the Q-switching pulse is in range between 75.4 and 96.2 mW and the pulse started to disappear at higher pumping. This is due to the SA, which continuously in the saturated state because of the high intensity of light, and there
is no time for de-excitation. Figure 5.15 (b) shows the measurement of the average output power and pulse energy at various pump power. The average output power increases from 5.3 to 10.9 mW as the pump power is increased from 75.4 to 96.2 mW. On the other hand, the pulse energy fluctuates within the pump power range. The maximum pulse energy of 252.6 nJ is obtained at 82.3 mW pump power.

Figure 5.15: The Q-switching performance of the laser (a) Repetition rate and pulse width, and (b) output power and pulse energy at various pump power
5.3.4 Mode-locking Laser Performances

As the output coupler ratio was changed to 90/10 in the YDFL cavity, the laser operation can be converted to mode-locking mode. The change of coupler has significantly reduced the cavity loss and with the help of nonlinear effect and dispersion characteristic inside the cavity, stable mode-locking pulses could be realized. The mode-locked operation also depended on the appropriate cavity length and SA optical properties.

The mode-locked YDFL has a total cavity length of around 9 m with a total net dispersion in normal regime of 0.2 ps². The self-started mode-locked operation was achieved at threshold pump power of 47.8 mW. Figure 5.16 summarizes the performance of mode-locking state of the fiber laser at 89.4 mW. The output spectrum is shown in Figure 5.16 (a) where no Kelly spectral sidebands was observed, indicating that the pulses operation of fiber laser is operating in a normal dispersion. It has a peak wavelength of 1072.7 nm and 3 dB bandwidth of 0.09 nm.

Figure 5.16 (b) shows the mode-locked pulse train with pulse repetition rate of 24.2 MHz and the pulses period (peak to peak spacing) of 41 ns which matches with the cavity length. This has confirmed that the operation of laser is in the mode-locking state and the operation was remaining stable until 89.4 mW pump power. As seen in the same figure, the pulse width is about 17.66 ns under the oscilloscope resolution and that is much broader than the real pulse width. As an alternative, the mathematical calculation based on time-bandwidth product (TBP) can be used for estimating the pulse width. As the TBP was assumed to be 0.441 for Gaussian pulse profile (Hisyam et al., 2017), the minimum possible pulse width is estimated to be approximately 18.8 ps.
Figure 5.16: The performances of the proposed mode-locked YDFL at pump power of 89.4 mW (a) Optical spectrum, (b) typical pulse train, and (c) The RF spectrum and (d) the output power and pulse energy characteristics against the pump power.

Figure 5.16 (c) shows the RF spectrum of the YDFL, which shows a SNR as high as 32 dB. It is worthy to note that no presence of fundamental frequency was observed when the SA is removed. Figure 5.16 (d) shows the output power and pulse energy at various pump powers. Both output power and pulse energy are linearly increased with increasing in pump power. At 89.4 mW pump power, the maximum output power and pulse energy is obtained to be around 18.6 mW and 0.8 nJ, respectively. The mode-locked pulse disappears when the pump power is increased above 89.4 mW.

5.4 Summary

Stable passively Q-switched YDFLs operating at 1057.4, and 1044.5 nm were successfully demonstrated using topological insulators of Bi$_2$Se$_3$ and Bi$_2$Te$_3$, respectively. Using Bi$_2$Se$_3$ saturable absorber, the YDFL obtains a stable train of pulses with a repetition rate frequency that is tunable from 48.83 to 102 kHz. Meanwhile, the corresponding pulse width decreases from 8.9 to 3.48 µs as the pump power increases.
from 67.6 to 88.3 mW. For the Bi$_2$Te$_3$ based Q-switched YDFL, the repetition rate grows from 43.25 to 93.63 kHz and the pulse width shrinks from 5.65 to 2.01 µs as the 980nm pump power rises from 72.8 to 98.4 mW. Both SAs show good Q-switching performance in the 1 µm wavelength region and the maximum pulse energy of 29.95 nJ was produced with the Bi$_2$Te$_3$ SA. A Sb$_2$Te$_3$ film was also demonstrated as an effective SA for generation Q-switching and mode-locking pulses in YDFL cavity. The threshold of generation Q-switched was obtained at 75.4 mW pump power and the laser remaining stable until pump power of 96.2 mW. The pulse repetition rate is increased from 24.4 to 55 kHz as the pump power increases from 75.4 to 96.2 mW. The pulse energy was around 252.6 nJ at 82.3 mW pump power, which is significantly higher than the Bi$_2$Te$_3$ based Q-switched laser. On the other hand, the mode-locking was also successfully achieved with Sb$_2$Te$_3$ SA by significantly reduce the cavity loss. The mode-locked fiber laser was obtained with 24.2 MHz repetition and 18.8 ps pulse width within the pump power between 47.8 to 89.4 mW. The maximum pulse energy was calculated to be around 0.8 nJ at at 89.4 mW pump power. These results verified that the Sb$_2$Te$_3$ is a better TI material as SA for Q-switching and mode-locking pulses generating in 1 micron wavelength region.
CHAPTER 6: TRANSITION METAL OXIDE NANOMATERIALS AS SATURABLE ABSORBER

6.1 Introduction

Very recently, a new kind of metal nanoparticles material such as gold, silver nanoparticles and titanium dioxide (TiO$_2$) were also investigated and demonstrated as a saturable absorber (SA) for Q-switched and mode-locked fiber lasers (Glubokov et al., 2014; T. Jiang et al., 2013). These types of SAs have gained attraction due to their unique properties such as proper modulation depth, low saturation intensity and fast response time. Previously, mode-locked fiber laser was demonstrated using gold nanoparticles based SA for generating 887 fs pulse train (X.-D. Wang et al., 2014). Silver nanoparticles was also proposed as SA for mode-locking and the absorption characteristic of the material was reported as 10 MW cm$^{-2}$ (Glubokov et al., 2014).

In this chapter, two types of metal oxide nanomaterials are explored as SA for Q-switching and mode-locking operations in Ytterbium-doped fiber laser (YDFL) cavity; Nickel Oxide (NiO) and cobalt oxide (Co$_3$O$_4$). These materials are embedded into a polymer film and sandwiched between two fiber ferrules, making it an SA device. By incorporating the all-fiber SA device into the YDFL cavity, Q-switching and mode-locking operations operating in 1 µm region could be realized.

6.2 Nickel Oxide Nanoparticles Thin Film based Q-switched and Mode-locked YDFL

Nickel oxide (NiO), a transition metal oxide semiconductor is an electro-chromic, transparent conducting, and antiferromagnetic material that have a wide range of applications in electro-chromic devices, optical fibers, and sensors. Nickel oxide nanoparticle could also be used as an effective SA since it has an appropriate modulation depth and low saturation intensity. The nonlinear refraction properties of NiO film was
also previously investigated using the thermally managed eclipse Z-scan technique (de Melo Jr et al., 2009). Many studies have also reported on NiO absorption properties where its band-gap energy is reported to be within a range from 3.6 to 4.0 eV, which equivalent to wavelength of 309 to 344 nm. However, Duan et. al reported that the nanoparticle absorption can be red-shifted by increasing the nanoparticle size (Duan et al., 2012). Therefore, NiO nanoparticle could be used as an SA in near-infrared region. Most recently, NiO film SA was successfully used to generate Q-switching pulses train in Erbium doped fiber laser (EDFL) cavity (Nady, Ahmed, Latiff, Numan, et al., 2017). However, so far, there are no reports on NiO based pulsed fiber laser operating in 1 µm region. Therefore, in this section, Q-switched and mode-locked fiber laser operating at 1 µm region are demonstrated using a NiO film as SA. The film material was sandwiched among two fiber ferrules, making it an SA device. By incorporating the NiO nanoparticle SA into the YDFL cavity, Q-switched and mode-locked operation could be achieved.

6.2.1 Fabrication and Characterization of Nickel Oxide Nanoparticles Thin Film SA

The fabrication process of the NiO nanoparticles thin film is described in Figure 6.1, which consist of two steps. In the first step, 1 M of NiCl$_2$·6H$_2$O was dissolved in 50 ml of deionized (DI) water under constant stirring for 10 min. After that, a freshly prepared solution of NaOH (2 M) was added dropwise to the above solution and consequently, subjected to sonication using a horn sonicator. The whole reaction was carried out using the constant amplitude of 60% for 1 h. The product was collected via centrifugation and washed several times with deionized water. Finally, the obtained sample was calcinated at 350 ºC for 3 hours in an electric box furnace. The synthesis of NiO nanoparticles details was reported in (Duraisamy et al., 2016).
In the second step, the NiO nanoparticles was mixed with the polyethylene oxide (PEO) as a host polymer to fabricate the thin film as described in Figure 6.1. The host polymer was prepared by dissolving 1 g of PEO in 120 ml of deionized water. Then, a suitable amount of NiO structure nanoparticles was added to the prepared solution and stirred it for further 2 hours. After that, the uniformly mixed slurry was cast on Teflon petri and dried at 60 ºC in a vacuum oven to fabricate the film. The thickness of the thin film was measured to be around 0.2 mm by using Mitutoyo micrometer screw gauge.

**Figure 6.1: The fabrication process of NiO nanoparticles thin film**

The X-ray diffraction (XRD) analysis pattern of the NiO nanostructure is shown in Figure 6.2 (a) and obviously indicates three peaks assigned to different vibrational modes. The three peaks are located at 37.28º, 43.28º, and 62.88º corresponding to crystalline NiO planes of (111), (200), and (220), respectively. The observed crystalline planes correspond to the face centered cubic structure of NiO. Compared to a Raman spectrum, the XRD is fit for indicating the phases as well as the crystalline orientation of the NiO. Figure 6.2 (b) shows the field emission scanning electron microscope (FESEM) image of NiO nanoparticles embedded into the PEO thin film. It has a high dense of agglomerated nanoparticles, which are formed as a thin film and randomly distributed on the surface.
These nanoparticles are in like-spherical shape with an average size of 35.78±8.67 nm. The nonlinear optical absorption properties of NiO thin film SA was also investigated using the twin-detector method as shown in Figure 6.3. For the NiO film sample that was used in this work, the modulation depth, non-saturable intensity, and saturation intensity were measured to be approximately 39 %, 49 %, and 0.04 MW/cm², respectively.

![XRD pattern and FESEM images of the NiO nanoparticles](image)

**Figure 6.2:** (a) XRD pattern and (b) FESEM images of the NiO nanoparticles embedded on the polymer thin film
6.2.2 The Fiber Laser Ring Configuration

The experimental setup of Q-switched YDFL based on NiO SA is shown in Figure 6.4. The optical ring cavity consists of a 1.5 m long Ytterbium-doped fiber (YDF), was forward pumped by a 980-nm laser diode (LD) through a 980/1064 nm wavelength division multiplexer (WDM). The YDF has a core and cladding diameters of 4 µm and 125 µm respectively, with a numerical aperture of 0.20, as well as the cut-off wavelength of 1010 nm, ion absorption of 280 dB/m at 920 nm and group velocity dispersion (GVD) of 24.22 ps²/km. An isolator was integrated into the optical ring cavity to ensure unidirectional operation. The NiO nanoparticles SA device was obtained by sandwiching a 1 mm × 1 mm piece of the film between two fiber ferrules using a fiber adaptor. Index matching gel was applied at the connection to minimize parasitic reflections. A 3 dB coupler was used to tap out the laser output. The output laser is analyzed by an optical spectrum analyzer with a spectral resolution of 0.02 nm, an optical power meter, and a 2
GHz photodetector together with a 350 MHz oscilloscope or 7.8 GHz radio-frequency (RF) spectrum analyzer.

The Q-switched laser was then converted into a mode-locked fiber laser by replacing the 50/50 output coupler with 90/10 coupler to reduce the cavity loss and allow the mode-locking pulses generation. The coupler allows 90% of the laser to oscillate in the cavity and only tap out 10% as an output. The mode-locked YDFL has a total cavity length of 9 m with a total net dispersion in normal regime of 0.2 ps$^2$, where the rest of the ring cavity is a single-mode fiber (HI 1060) with a GVD of 21.91 ps$^2$/km.

Figure 6.4: The schematic diagram of experimental setup for Q-switched and mode-locked YDFLs with NiO based SA
6.2.3  Result and Discussion for the Q-Switching Operation

The self-starting Q-switched YDFL was obtained at threshold pump power of 117.73 mW and its operation was maintained up to pump power of 133 mW. Figure 6.5 (a) shows the optical spectrum of Q-switched YDFL when the pump power is fixed at 117.73 mW. As seen, the spectrum is centered at 1073.5 nm with a bandwidth of 0.6 nm. Figure 6.5 (b) shows the average output power against the pump power. The output power increased from 3.3 to 7.0 mW when the pump power increased from 117.73 to 133 mW.

Figure 6.5: (a) Output spectrum at the threshold pump power of 117.73 mW, (b) Output power against pump power
Figure 6.6 (a) shows the Q-switched YDFL pulse trains at three different pump powers of 117.73, 125.4, and 133 mW, respectively. As the pump power increases, more energy can be stored in the laser cavity and this contributes to the rise in the repetition rate accompanied by the reduction in pulse duration. The repetition rates have the period of 9.5, 12, and 15.8 kHz, while the corresponding pulse durations are 14.27, 11.28, and 10.18 μs at pump power of 117.73, 125.4, and 133 mW, respectively. The properties of the Q-switching laser principally rely on upon the gain medium, SA and pump power. Figure 6.6 (b) shows RF spectrum of the Q-switched YDFL at pump power of 133 mW, for which the pulse repetition rate is 15.8 kHz, matching with the oscilloscope data. The RF signal-to-noise ratio (SNR) is about 30 dB with no spectral modulation, indicating that the Q-switching operation is stable.

Figure 6.6 (c) shows the repetition rate and pulse width as a function of 980 nm pump power. At the point when the pump power was expanded from 117.73 to 133 mW, the repetition rate increased in a practically direct manner from 9.5 to 15.8 kHz, while the pulse width gets to be smaller, shortening from 14.27 to 10.18 μs. This is an average normal for Q-switching operation. As pump power increases, more power flows inside the laser cavity, in this manner saturating the SA faster. It can be seen that the SA become almost saturated because of the smaller changes of repetition rate, and pulse width. This appearance can be linked into the modulation depth and the length of the laser cavity. Figure 6.7 shows the variation of peak power and pulse energy against the pump power. As the pump power increased from 117.73 to 133 mW, the peak power was also increased from 22.9 to 40.9 mW. The pulse energy maximum pulse energy is 478 nJ at the same range of pump power, which the measuring based on the repetition rate and output power. The peak power is depended on the pulse energy and pulse duration.
Figure 6.6: Temporal performance of Q-switched YDFL (a) Pulse train at different pump power. (b) RF spectra at pump power of 133 mW with 150 kHz span. Insert is enlarged image of 15.8 kHz repetition rate. (c) Repetition rate and pulse with respect to pump power.
6.2.4 Result and Discussion for the Mode-locking Operation

By changing the coupler to 90/10 coupler in the ring cavity, the cavity loss significantly reduces which support the generation of a stable mode-locking pulse. In the
experiment, the continuous-wave laser threshold pump power is approximately 103 mW. A self-started mode-locked was achieved at a pump power of 110 mW until 137.5 mW. The generation of mode-locking operation is completely depended on the SA optical properties and suitable cavity length. Figure 6.8 (a) shows the optical spectrum of the mode-locking pulse at a pump power of 137.5 mW with the peak wavelength of 1037.72 nm and 3 dB spectral bandwidth of 0.08 nm with no Kelly sideband. Thus, it confirmed the pulses operation in the normal dispersion. Figure 6.8 (b) displays the mode-locked YDFL pulse train, which corresponded to 23 MHz repetition rate with the optical cavity length of 9 m and 49 ns pulse period (peak to peak space) while remaining stable until 137.5 mW pump power. As shown in the same figure with the limitation of the oscilloscope resolution, the pulse width is 25 ns, which is much broader than the actual pulse width. As an alternative, the mathematical calculation can be used for measuring the pulse width based on time-bandwidth product (TBP). Assuming the TBP of 0.441 for Gaussian pulse profile (Hisyam et al., 2017), the minimum possible pulse width is mathematically estimated as approximately 19.8 ps. The mode-locking pulse train was observed to run stably at room temperature. Figure 6.8 (c) shows clearly the radio frequency spectrum of the mode-locking operation, which has a SNR up to 29 dB. Once NiO-SA removed from the fiber ferrule, the obtained fundamental frequency will terminate.

The relationship between the output power and input pump power is shown in Figure 6.8 (d), which the maximum output power is about 12.8 mW at 137.5 mW pump power. Overall performance of mode-locked YDFL has a slope efficiency of 38.22%. Figure 6.8 (e) shows the relationship of mode-locked YDFL between pulse energy and peak power against pump powers. Both pulse energy and peak power is linearly increased against pump power. The maximum pulse energy and peak power are 0.56 nJ and 26.4 W,
respectively, at 137.5 mW pump power. The mode-locked pulse disappears when the pump power is increased above 137.5 mW.

Figure 6.8: Mode-locked performance. (a) The optical spectrum, (b) pulse train (insert the image of single-envelope pulse), (c) the RF spectrum at maximum pump power of 137.5 mW. (d) Output power, and (e) Pulse energy and peak power against the pump power.
Figure 6.8, continued

(c) [Graph showing intensity vs. frequency with a peak at 29 dB]

(d) [Graph showing output power vs. pump power with a linear relationship]

Figure 6.8, continued
6.3 Cobalt Oxide Film SA for Generating Q-Switching and Mode-locking Pulses in YDFL cavity

Very recently, transition metal oxides such as titanium dioxide (TiO$_2$) and zinc oxide (ZnO) has used as SAs, which have been reported to generate an ultra-short pulse fiber laser (H Ahmad, CSJ Lee, et al., 2016; H Ahmad, Siti Aisyah Reduan, et al., 2016). In the previous section, NiO nanoparticles which were embedded into PEO polymer was demonstrated as SA for operation in YDFL cavity. The transition metal oxides have gained more attentions among many researchers due to their large optical nonlinearity and the advantages of good thermal and the chemical stability as well as the mechanical strength. Among those materials, cobalt (Co$_3$O$_4$) has the best ratio performance of the nonlinearity to the linear absorption. It is also a promising material for fabricating highly...
nonlinear transmission devices in the visible range. The nonlinear refraction properties of Co$_3$O$_4$ film was previously reported (Ando et al., 2004; Yamamoto et al., 2002; X Zhu et al., 2010; Xiushan Zhu et al., 2012). It has a bandgap size at 1.38 eV and 2.0 eV, which corresponds to 898 nm and 620 nm, respectively. This makes the material as promising candidates for fabricating highly nonlinear transmission devices in the visible range. In addition, Barecca et al. (2001) reported that the absorption also presence at 1-micron region, due to internal oxidation-reduction process that caused inter-valence charge transfer from Co(II) to Co(III) (Barreca et al., 2001). The first successful generation of Q-switched fiber laser using Co$_3$O$_4$ film SA was demonstrated at 1.55-micron region (Nady, Ahmed, Numan, et al., 2017). It generates stable pulse up to 70.92 kHz with a signal-to-noise ratio (SNR) of 60 dB. However, no scientific report has been found for 1-micron region.

In this section, a Q-switched and mode-locked ytterbium-doped fiber laser (YDFL) is reported based on Co$_3$O$_4$ nanocubes thin film as SA. The Co$_3$O$_4$ nanocubes were embedded in PEO film, which was then integrated into YDFL cavity. A stable Q-switched laser operating at 1043.64 nm wavelength was obtained and the pulse repetition rates are tunable from 60.3 to 86.66 kHz as the pump power was varied from 144.4 to 165.4 mW. The mode-locked laser operated at 1035.8 nm wavelength with 20 MHz repetition rate. The maximum average output power and pulse energy were 24 mW and 1.2 nJ respectively at pump power of 186 mW. This is the first reporting of Co$_3$O$_4$ nanocubes thin film as SA used in a mode-locked laser.

6.3.1 Fabrication and Characterization of Co$_3$O$_4$ Film SA

The Co$_3$O$_4$ nanocubes used in this study was synthesised via a hydrothermal method as described in Figure 6.9. In the first step, 1 mmole of Co(CH$_3$COO)$_2$: 4H$_2$O solution was mixed with 45 ml of deionized (DI) water and ethanol (3:1) mixture then sonicated
by ultrasonic bath. Next, the mixture was put together with Carbonyldiamide, as commonly called urea solution, of 15 ml followed by stirring for 30 minutes. After that, the stirring mixture was put in a 100 mL Teflon-lined stainless steel autoclave and subjected to hydrothermal treatment by keeping the temperature of the oven at 150 °C for 5 hours. Finally, the synthesized Co$_3$O$_4$ nanocubes was washed several times by DI water and ethanol under centrifugal force and dried in a hot vacuum oven at 60 °C for 24 hours.

For practical use of Co$_3$O$_4$ nanocubes as SA, the Co$_3$O$_4$ nanocubes were embedded into PEO to form thin film via solution casting route as described in the second step of Figure 6.9. Firstly, 1 g of PEO solution was prepared as a diluted solution by dissolving in 120 mL of DI water and then stirring with continuously stirring at 50°C. This diluted solution was kept under continuously stirring for 2 hours. After that, some amount of the synthesized Co$_3$O$_4$ nanocubes was mixed to the diluted solution, followed by stirring for 2 hours to form a uniform solution mixture as a mixed slurry. Finally, the mixed slurry was casted to Teflon coated Aluminum foil to form thin film followed by drying in the oven at 60 °C.

For characterization studies, FESEM image was used to obtain the surface morphology of the synthesized Co$_3$O$_4$ nanocubes. For higher magnification, high resolution transmission electron microscopy (HRTEM) was obtained by JEOL JEM-2100F. For analytical structure, micro-Raman scattering spectra was used to investigate the synthesized Co$_3$O$_4$ nanocubes structure using an argon-ion laser with excitation wavelength of 514 nm and XRD for structural crystallinity and purity of the Co$_3$O$_4$ nanocubes was obtained by Philips X’pert X-ray diffractometer using copper Ka radiation with a wavelength of 1.5406 Å for a scan rate of 0.02 degree per second. Mitutoyo micrometer screw gauge measurement was used to determine the thin film thickness which was about 0.05 mm.
The surface morphology of the synthesized Co$_3$O$_4$ nanocubes by hydrothermal method is shown in Figure 6.10 (a). The FESEM image presented nanocubes form with high density that appeared in agglomeration forms. Figure 6.10 (b) shows the synthesized Co$_3$O$_4$ nanocubes at higher magnification presented by HRTEM image. Furthermore, the nanocubes approximately appeared in uniformity size with agglomerations which the average size was around 32 nm. Figure 6.11 (a) shows the Raman scattering spectrum of the nanocubes grown by hydrothermal method. This spectra consists of five modes at around 192, 482, 517, 614 and 687 cm$^{-1}$ which can be attributed to the B$_{1g}$, E$_g$, F$_{1g}$, F$_{2g}$ and A$_{1g}$ modes of Co$_3$O$_4$, respectively (Numan et al., 2017) and that confirm the appeared of nanocubes at FESEM image belong to Co$_3$O$_4$ nanocubes structure. XRD patterns of the Co$_3$O$_4$ nanocubes is shown in Figure 6.11 (b) and mainly presented seven planes of the crystallite of the nanocubes. The crystalline peaks are located at 18.1°, 30.9°, 36.4°, 44.2°, 54.9°, 58.6°, and 64.4° corresponding to crystalline Co$_3$O$_4$ planes of (111), (220), (311), (400), (422), (511), and (440), respectively (Numan et al., 2016; Liangmiao Zhang
et al., 2013). The nonlinear optical absorption properties of Co$_3$O$_4$ thin film SA was achieved using twin-detector method in order to confirm its saturable absorption as shown in Figure 6.12. For the Co$_3$O$_4$ SA sample that was used in this work, the modulation depth and saturation intensity were measured to be approximately 0.35 % and 3 MW/cm$^2$, respectively, which few materials as SAs have also a small modulation depth (Junsu Lee et al., 2015; Z. Luo et al., 2014).

Figure 6.10: The images of Co$_3$O$_4$ samples (a) FESEM, and (b) HRTEM

Figure 6.11: (a) Raman spectra and (b) XRD pattern of Raman spectrum of the Co$_3$O$_4$ nanocubes
6.3.2 Configuration of Co$_3$O$_4$ based Q-switched and Mode-locked YDFL

In the experiment, the application of Co$_3$O$_4$ nanocubes film as a SA in generating mode-locking pulses train in YDFL was investigated. The experimental setup is shown in Figure 6.13, which mainly consists of 1.5 m long YDF as a gain medium. In order to
prevent the laser reflecting, an isolator is spliced into the cavity. The 980 nm pump is coupled into the fiber ring resonator with a 980/1064 nm wavelength division multiplexer (WDM). The Q-switched laser was obtained by using a 3dB output coupler inside the cavity. A 90/10 output coupler was used in the mode-locked laser cavity to extract 10% of the output for analysis. The remainder of laser power was channeled back to the SA device and kept in the cavity to oscillate for laser generation. The Co$_3$O$_4$ nanocubes film SA was constructed by sandwiching a ~ 1 mm×1 mm piece of the film between two fiber ferrules using a fiber adaptor while applying index matching gel at the connection to minimize parasitic reflections. The output laser was routed to an OSA and optical power meter for spectral and output power analysis, respectively. It is also measured by a 2 GHz photodetector, which was connected to a 350 MHz oscilloscope and 7.8 GHz RF spectrum analyzer for temporal and frequency analysis, respectively. The mode-locked YDFL has a total cavity length of 10 m with a total net dispersion in normal regime of 0.22 ps$^2$.

Figure 6.13: The employment of Co$_3$O$_4$ nanocubes film based SA for generating Q-switching and mode-locking pulses trains in YDFL cavity. Q-switched and mode-locked lasers employ 50/50 and 90/10 output coupler, respectively
6.3.3 Q-Switching Performance of the Co$_3$O$_4$ based YDFL

In this experiment, a self-starting Q-switching pulse was generated in the Co$_3$O$_4$ based YDFL cavity at threshold pump power of 144.4 mW. Figure 6.14 shows the optical spectrum of the proposal Q-switched YDFL at the threshold pump power. As shown in the figure, the spectrum centered at wavelength of 1043.64 nm with bandwidth of 0.24 nm. The stable Q-switching pulse trains were maintained up to the pump power of 165.4 mW. The pulses became unstable as the pump power further increased. Figure 6.15 (a)-(c) shows the Q-switching pulse trains with no timing jitter noticeable and its typical pulse envelope at three different pump powers of 144.4, 155, 165.4 mW. The pulse train has the repetition rates of 60.3, 78.13, and 86.66 kHz, which corresponds to the periods of 16.6, 12.8, and 11.5 µs at pump powers of 144.4, 155, 165.4 mW, respectively. The single pulse profile has the symmetrical Gaussian-like shape with full-width half maximum (FWHM) of 4.4, 5, and 4.6 µs at the same three different pump powers, respectively.

![Output spectrum of the proposed Q-switched YDFL with Cobalt SA at the threshold pump power of 144.4 mW](image_url)
Figure 6.15: Q-switched YDF of pulse train and single-pulse envelope at different pump powers (a) 144.4 mW (repetition rate of 60.3 kHz), (b) 155 mW (repetition rate of 78.13 kHz), and (c) 165.4 mW (repetition rate of 86.66 kHz)
Figure 6.16 shows the characteristics of repetition rate and output power at various 980 nm pump power. Both repetition rate and output power increases with the pump power as shown at the same figure. The pulse repetition rate increases from 60.3 to 86.66 kHz as the pump power is varied from 144.4 to 165.4 mW, which confirm the Q-switching operation of the laser. The repetition rate of Q-switching pulse is able to change due to the lifetime of gain medium and the pump power. On the other hand, the output power linearly increases from 8.9 to 13.2 mW as the pump power is increased from 144.4 to 165.4 mW. This can be related to the ytterbium ion in the gain medium, which excite more electrons to the higher level with more pump power. The $\text{Co}_3\text{O}_4$ nanocubes film SA is saturated faster with higher pump power and thus increases the repetition rate.

Figure 6.17 shows the pulse width and pulse energy as function of pump power. It is observed that both pulse width and pulse energy are almost unchanged as the pump power is increased from 144.4 to 165.4 mW. The pulse width fluctuates within 4 to 5.16 µs with the shortest pulse width is obtained at 147.9 mW pump. The maximum pulse energy of 154 nJ is achieved at pump power of 147.9 mW. The pulse energy could be improved by reducing the insertion loss of the SA or by optimizing the laser cavity. As the pump power increases, the non-radiative decay rate of the $\text{Yb}^{3+}$ ions also increases. This generates heat in the laser cavity system, which it’s absorbed by the SA. This will energize some electrons in the SA’s valance band to move up to the conduction band from the strong internal thermal motion. As fewer electrons in the valence band are available for photon absorption, the initial transmittance of the SA becomes greater as the pump power or the levels of absorber heat increases. Thus, the SA’s efficiency as SA is greatly reduced, meaning that the pulse width begins to increase again after the saturation of SA. The pulse started to disappear at higher pumping of 165.4 mW due to the SA, which is continuously in the saturated state.
Figure 6.16: The characteristics of repetition rate and output power at various pump power

Figure 6.17: The characteristics of pulse width and pulse energy at various pump power
6.3.4 Mode-locking Performance of the Co₃O₄ based YDFL

For the YDFL cavity with 90/10 output coupler, the laser started to laser in CW mode at the threshold pump power of 117 mW. As the pump power was increased gradually to 123.7 mW, the self-starting mode-locking was achieved and its operation was maintained up to 186 mW pump power. The generation of mode-locking pulses was completely depended on the SA optical properties and suitable cavity length. Figure 6.18 (a) shows the output spectrum of the mode-locked YDFL at pump power of 186 mW. It operates at center wavelength of 1035.8 nm with a 3dB spectral bandwidth of 0.32 nm. There was no Kelly sideband observed in the spectrum, which confirmed the normal dispersion operation of the mode-locked laser. The narrow spectral bandwidth was due to the high net cavity dispersion and minimal nonlinearity induced by the high pump power (Sathiyan et al., 2016).

The typical oscilloscope trace of the proposed mode-locked YDFL is shown in Figure 6.18 (b). It is shown that the measured repetition rate of the pulse is approximately 20 MHz, which is matched with the cavity length of 10 m and the pulse period (peak to peak space) of 51 ns. The oscilloscope trace displays a 24 ns pulse width. But, due to the limitation of the oscilloscope resolution, the actual pulse width is much smaller than the value. Since an auto-correlator was not available for 1µm, the actual pulse width was estimated based on mathematical calculation. Assuming the pulse shape follows the Gaussian profile, the TBP should be 0.441 (Hisyam et al., 2017), thus the minimum possible pulse width was estimated to be approximately 4.93 ps. Figure 6.18 (c) shows the RF spectrum for the mode-locked YDFL which showing two modes within 50 MHz frequency range. The SNR of the fundamental frequency was 37 dB, which indicates the stability of the mode-locking pulses.
Figure 6.18: Mode-locking performance of the Co$_3$O$_4$ based YDFL (a) The optical spectrum, (b) pulse train (insert the image of single-envelope pulse) and (c) the RF spectrum, at the maximum pump power of 186 mW
Figure 6.18, continued

Figure 6.19: (a) Output power and pulse energy, and (b) Peak power against the pump power
Figure 6.19, continued

Figure 6.19 shows the output power, pulse energy, and the peak power against pump powers. All the output power, pulse energy and peak power show an increasing trend with the pump power. The first lasing was a continuous wave, which its output power was approximately 1.67 mW at pump power of 117 mW. The CW laser was converted into a mode-locked laser at pump power of 123.7 mW. The output power was linearly increased from 2.8 to 24 mW when the pump power increased from 123.7 to 186 mW as shown in Figure 6.19 (a). The maximum pulse energy and peak power are approximated 1.2 nJ and 228.7 W at 186 mW pump power shown in Figures 6.19 (a) and (b), respectively. It is worthy to note that the mode-locking pulses train disappears when the pump power is increased above 186 mW. This is due to the SA, which continuously stay in the saturated state because of the high intensity of light.
6.4 Summary

Passive Q-switched and mode-locked YDFLs were successfully demonstrated based on two types of metal oxide nanomaterials; NiO nanoparticles and Co$_3$O$_4$ nanocubes thin film SAs. With NiO nanoparticles film SA, a stable self-starting Q-switched laser was achieved in the YDFL cavity configured with 1.5m long YDF as the pump power is increased above 117.73 mW. The pulses have the maximum pulse energy of 478 nJ, shortest pulse duration of 10.18 μs and repetition rates are tunable from 9.5 to 15.8 kHz. It is also found that the mode-locking pulses can be realized in the cavity by changing the output coupler to 90/10 coupler instead of 3 dB coupler. The mode-locking pulses train was obtained at pump power range from 97.3 mW to 137.5 mW. The mode-locked laser operated at 1037.72 nm wavelength with a fixed repetition rate of 23 MHz and picoseCONDS pulse width. The maximum pulse energy and peak power were calculated to be around 0.56 nJ and 26.4 W, respectively at 137.5 mW pump power.

Stable Q-switched and mode-locked YDFLs were realized with Co$_3$O$_4$ nanocubes SA, which was obtained by embedding the nanocubes into PEO film. The Q-switched laser operated at 1043.64 nm wavelength with the pulse repetition rate obtained varies from 60.3 to 86.66 kHz by increasing the pump power from 144.4 to 165.4 mW. The shortest pulse width and highest pulse energy were 4 μs and 154 nJ, respectively. The mode-locked Co$_3$O$_4$ based YDFL was also realized within a pump power range from 123.7 mW to 186 mW by utilizing a 90/10 output coupler inside the cavity. The mode-locked laser operated at 1035.8 nm wavelength with a fixed repetition rate of 20 MHz and picoseconds pulse width. The maximum pulse energy and output power were calculated to be around 1.2 nJ and 24 mW, respectively at 186 mW pump power. These results show that both NiO nanoparticles and Co$_3$O$_4$ nanocubes nanomaterials can be engaged for Q-switched and mode-locked fiber laser applications in 1 μm wavelength region.
CHAPTER 7: CONCLUSION AND FUTURE WORKS

7.1 Conclusions

Fiber laser has a broad range of applications ranging from industry to optical communication. Pulsed fiber laser with high peak intensity and high pulse energy is suitable for micromachining and drilling which benefits the medical, electronic and automotive industry because of its simplicity and compactness. Lasers use the quantum impact of excited emission to produce light and they are constructed based on the following elements; an active medium as a gain provider, a pumping source to produce the energy and an optical cavity to reinforce and control the optical field. Ytterbium-doped fiber laser (YDFL) is the most efficient fiber lasers used as a laser active dopant in the form of the trivalent ion Yb$^{3+}$ to generate laser in 1 micron region, which has many applications in various areas. The Ytterbium-doped fiber (YDF) is an attractive gain medium since it has a simple energy level structure, high reliability, compact and the lasing wavelength at 1 µm, within the low-loss of optical laser as well as it is capable for producing high output power.

Q-switched or mode-locked fiber lasers can be generated by either active or passive techniques. The active technique is usually required an additional switching electronics device such as acousto-optic modulators to be incorporated in the laser cavity. Compared with active technique, passive technique is preferable for generation Q-switching and mode-locking pulse in fiber laser due to their advantages such as nature being compact, low cost, flexibility and easy to setup. Passive technique is normally demonstrated by utilizing saturable absorption of optical materials. The main objectives of this research work was to fabricate and demonstrate an efficient and low-cost Q-switched and mode-locked YDFLs operating in 1 µm region by utilizing new nanomaterials as SA.
In the chapter 3, molybdenum disulfide (MoS$_2$) was proposed as SA for generating Q-switched and mode-locked pulses train in YDFL cavity. MoS$_2$ is drawing more interest for Q-switching and mode-locked fiber lasers applications. The reason for this growing interest is due to its material’s thickness, which gives an excellent optical laser properties as well as the band-gap. The passively Q-switched YDFL was demonstrated by using a few layer MoS$_2$, which was mechanically exfoliated from a natural MoS$_2$ crystal. The mechanical exfoliation was done by using a scotch tape and the acquired MoS$_2$ layers was sandwiched between two fiber ferrules to form a fiber compatible Q-switcher. By introducing the MoS$_2$ SA into the YDFL cavity, a stable pulse laser is generated at 1070.2 nm wavelength with repetition rate is tunable from 3.817 to 25.25 kHz. A passively mode-locked YDFL was also demonstrated using a MoS$_2$ polymer film which was prepared using liquid phase exfoliation approach as a SA. The laser was successfully generating a mode-locking pulses with repetition rate of 18.8 MHz and pulse energy of 0.1 nJ. The LPE method was simple, cheap and does not require any complicated procedures.

In chapter 4, another 2D material: Black phosphorus (BP) is proposed as new SA for operation at 1 micron region. Both Q-switching and mode-locking operations were demonstrated using a YDF as a gain medium. BP dubbed as the newly emerging allotrope, has gained much attention for potential applications in the next-generation optoelectronics devices such as sensor, field-effect transistors and solar cells. The BP comprises only the elemental “phosphorus” and thus it could be easily peeled off by mechanical exfoliation. At first, a Q-switched YDFL was obtained by using a multi-layer BP SA, which was prepared by fixing the BP material onto a sticky tape using a mechanical exfoliation technique. The laser has a pump threshold of 55.1 mW, a pulse repetition rate that was tunable from 8.2 to 32.9 kHz, and the narrowest pulse width of 10.8 µs. The highest pulse energy of 328 nJ was achieved at the pump power of 97.6 mW. In mode-locking experiment, the BP SA was prepared using a slightly different method
to reduce the device loss and thus enable the mode-locking operation. The SA was prepared by mechanically exfoliation of the BP crystal and fixed the acquired flakes onto the end surface of a standard FC/PC fiber connector. By incorporating the BP SA into the YDF laser cavity, a stable mode-locked operation was obtained at 1033.76 nm wavelength and the repetition rate was 10 MHz. These experimental results indicate that the BP SA can be used successfully to generate stable Q-switching and mode-locking pulses train at 1 µm region.

In chapter 5, passively Q-switched YDFLs were demonstrated using a few-layers Bi$_2$Se$_3$, Bi$_2$Te$_3$ and antimony telluride (Sb$_2$Te$_3$) based SAs while mode-locked YDFL was demonstrated using Sb$_2$Te$_3$ material to exploit the wideband saturable-absorption characteristic of the Topological insulators (TIs). TIs have attracted attention of researchers due to their high nonlinear susceptibility and large modulation depth with efficient saturable absorption property. They are excellent semiconductors with high surface electron mobility. For this purpose, free-standing Bi$_2$Se$_3$, Bi$_2$Te$_3$- and Sb$_2$Te$_3$-polymer composite SA films were prepared by implanting the powder of these materials into a polyvinyl alcohol (PVA) host. These films are used in a fully fiber-integrated laser cavity for generating pulses train.

Using Bi$_2$Se$_3$ saturable absorber, the Q-switched YDFL obtains a stable train of pulses with a repetition rate frequency that is tunable from 48.83 to 102 kH while the corresponding pulse width decreases from 8.9 to 3.48 µs as the pump power increases from 67.6 to 88.3 mW. For the Bi$_2$Te$_3$ based Q-switched YDFL, the repetition rate grows from 43.25 to 93.63 kHz and the pulse width shrinks from 5.65 to 2.01 µs as the 980nm pump power rises from 72.8 to 98.4 mW. The Sb$_2$Te$_3$ film based Q-switched YDFL produced pulse repetition rate, which is increased from 24.4 to 55 kHz as the pump power increases from 75.4 to 96.2 mW. The pulse energy was around 252.6 nJ at 82.3 mW pump
power, which is significantly higher than both Bi$_2$Se$_3$ and Bi$_2$Te$_3$ based Q-switched lasers. On the other hand, the mode-locking pulses was also successfully achieved with Bi$_2$Te$_3$ SA. It operates at 24.2 MHz repetition and 18.8 ps pulse width within the pump power between 47.8 to 89.4 mW. The maximum pulse energy was calculated to be around 0.8 nJ at at 89.4 mW pump power.

Two types of transition metal oxide nanomaterials were also explored as SA for Q-switching and mode-locking operations in YDFL cavity as described in Chapter 6. These materials were Nickel Oxide (NiO) and cobalt oxide (Co$_3$O$_4$), which were embedded into a polymer film to realize Q-switching and mode-locking pulses operating in 1 µm region. With NiO nanoparticles film SA, a stable self-starting Q-switched laser was achieved with the maximum pulse energy of 478 nJ, shortest pulse duration of 10.18 µs and repetition rates were tunable from 9.5 to 15.8 kHz. It is also found that the mode-locking pulses can be realized in the cavity by changing the output coupler to 90/10 coupler instead of 3 dB coupler. The mode-locked laser operated at 1037.72 nm wavelength with a fixed repetition rate of 23 MHz and picoseCONDS pulse width. Stable Q-switched and mode-locked YDFLs were realized with Co$_3$O$_4$ nanocubes SA. The Q-switched laser operated at 1043.64 nm wavelength with the pulse repetition rate obtained varies from 60.3 to 86.66 kHz by increasing the pump power from 144.4 to 165.4 mW. The mode-locked Co$_3$O$_4$ based YDFL was also realized within a pump power range from 123.7 mW to 186 mW by utilizing a 90/10 output coupler inside the cavity. The mode-locked laser operated at 1035.8 nm wavelength with a fixed repetition rate of 20 MHz and picoseconds pulse width.
Table 7.1: Summary of the SAs performances.

<table>
<thead>
<tr>
<th>SA</th>
<th>Repetition rate (kHz)</th>
<th>Min pulse duration (µs)</th>
<th>Max pulse energy (nJ)</th>
<th>Modulation depth (%)</th>
<th>Saturation intensity (MW/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MoS₂</td>
<td>3.817-25.25</td>
<td>11.33</td>
<td>295.45</td>
<td>11.3</td>
<td>23.5</td>
</tr>
<tr>
<td>BP</td>
<td>8.1-32.9</td>
<td>10.8</td>
<td>328</td>
<td>7</td>
<td>0.0025</td>
</tr>
<tr>
<td>Bi₂Se₃</td>
<td>48.83-102</td>
<td>3.48</td>
<td>16.6</td>
<td>15.9</td>
<td>50</td>
</tr>
<tr>
<td>Bi₂Te₃</td>
<td>72.8-98.4</td>
<td>2.01</td>
<td>29.95</td>
<td>30</td>
<td>40</td>
</tr>
<tr>
<td>Sb₂Te₃</td>
<td>24.4-55</td>
<td>6.6</td>
<td>252.6</td>
<td>8.5</td>
<td>36</td>
</tr>
<tr>
<td>NiO NPs</td>
<td>9.5-15.8</td>
<td>10.18</td>
<td>478</td>
<td>39</td>
<td>0.04</td>
</tr>
<tr>
<td>Co₃O₄ nanocubes</td>
<td>60.3-86.66</td>
<td>4</td>
<td>154</td>
<td>0.35</td>
<td>3</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>SA</th>
<th>Repetition rate (MHz)</th>
<th>Pulse duration (ps)</th>
<th>Max pulse energy (nJ)</th>
<th>Modulation depth (%)</th>
<th>Saturation intensity (MW/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MoS₂-PVA</td>
<td>18.8</td>
<td>28.3</td>
<td>0.1</td>
<td>9.7</td>
<td>40</td>
</tr>
<tr>
<td>BP</td>
<td>10</td>
<td>3.27</td>
<td>2.7</td>
<td>7.5</td>
<td>0.35</td>
</tr>
<tr>
<td>Sb₂Te₃</td>
<td>24.2</td>
<td>18.8</td>
<td>0.8</td>
<td>8.5</td>
<td>36</td>
</tr>
<tr>
<td>NiO NPs</td>
<td>23</td>
<td>19.8</td>
<td>0.56</td>
<td>39</td>
<td>0.04</td>
</tr>
<tr>
<td>Co₃O₄ nanocubes</td>
<td>20</td>
<td>4.93</td>
<td>1.2</td>
<td>0.35</td>
<td>3</td>
</tr>
</tbody>
</table>

In conclusion, several types of nanomaterials based SAs such as, Molybdenum disulfide (MoS₂), Black Phosphorus (BP), Topological Insulator (TI), Nickle Oxide (NiO) nanoparticles, and Cobalt oxide (Co₃O₄) nanocubes have been fabricated and characterized. It is obtained that all the fabricated SAs are capable to operate in YDFL cavity for generating Q-switching pulses with high energy. The generation of the mode-locking pulses in YDFL cavity have also been obtained using the fabricated nanomaterials as SA. These nanomaterials based SA could be useful as a simple, low cost, and low-loss
for operation in 1 µm region. Table 7.1 summarizes the performances of those SAs. Each SA has different laser performance and this laser performance can be applied for specific application based on pulse width, pulse energy and peak power. From the summary table we can see that BP produced shorter pulse width, where NiO has higher pulse energy.

7.2 Future Works

This section provides diverse interesting directions for the future work. The future work should focus on exploring the applications of the developed SAs in other wavelength regions such as 1.5, 2 and 3.5 micron using an Erbium-doped fiber (EDF), thulium-doped fiber and ZBLAN EDF as the gain medium, respectively. Further work should also focus on improving the current 1 µm region lasing characteristics and ultra-short pulse generation. The performance of Q-switching and mode-locking pulses state can also be realized and improved by using other types of nanomaterials such as gold, and silver nanoparticles as SAs. The mode-locking pulse generation can also be improved by utilizing more laser configurations with a better controlling of cavity nonlinear effect and dispersion. The main focus of investigation is by replacing the gain medium with higher dopant active fiber that can keep a high gain per length of the lasers. The other key focus for future work is to investigate a dual wavelength or multi-wavelengths pulse fiber laser using a special configuration such as adding PCF or side-polished fiber into the laser cavity. For example, Q-switched multi-wavelength fiber laser maintains good possibility applications such as optoelectronics, sensors, communications and instrumentations.
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Below is the list of publications and conference during the study;


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Q-switched Ytterbium doped fiber laser using a multi-walled carbon nanotubes embedded in polyethylene oxide film

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We demonstrate a passively Q-switched Ytterbium-doped fiber laser (YDFL) using a multi-walled carbon nanotubes (MWCNTs) embedded in polyvinyl alcohol (PVA) film as a passive saturable absorber (SA). The SA film is prepared by mixing the MWCNTs homogeneous solution with a dilute PVA polymer solution. It is sandwiched between two FC/PC fiber connectors and integrated into the laser cavity to generate a stable Q-switching pulse operating at wavelength of 1058.6 nm with a threshold pump power of 65.72 mW. The repetition rates of the laser varies from 5.484 kHz to 32.19 kHz as the pump power increases from 65.72 mW to 97.29 mW. The lowest pulse width of 4.2 μs is obtained at the pump power of 69.71 mW while the highest pulse energy of 211.8 nJ is obtained at pump power of 77.75 mW.

(Received August 13, 2015; accepted September 9, 2015)

Keywords: Ytterbium-doped fiber laser, Multi-walled carbon nanotubes/polyvinyl alcohol film, Q-switching, passive Q-switched

1. Introduction

Passively Q-switched fiber lasers with the advantages of compactness and flexibility have attracted much attention in recent years due to their potential applications in laser material processing, remote sensing, telecommunications and medicine [1-4]. They are normally obtained using active approaches by inserting an acoustic-optic or an electro-optic modulator into the laser cavity to generate a short optical pulse through sudden switching of the cavity Q-factor. Recently, passive methods are also widely explored for Q-switching in fiber lasers by utilizing the saturable absorption of optical materials (i.e. saturable absorbers, SAs). In the past, several kinds of SAs such as semiconductor saturable absorber mirrors (SESAMs) [1], carbon nanotubes (CNTs) [2], graphene [3-4] have been successfully used to passively generate Q-switch fiber lasers. Although SESAMs have been well developed and commercially available for the past two decades, they exhibit obvious disadvantages such as complex fabrication, expensive packaging and low damage threshold. The CNTs and graphene absorbers are simpler in fabrication and cheaper than SESAMs, and they operate in a wider wavelength band with a shorter recovery time.

To date, many works have been reported on the use of single-walled carbon nanotubes (SWCNTs) to generate passive Q-switching [2] and mode-locking pulse trains [5]. Recently, a new member of CNTs family, multi-walled carbon nanotubes (MWCNTs) have also drawn the attention of researchers due to their many advantages [6-7]. For instance, the MWCNTs material does not need complicated techniques or special conditions to grow such that its production yield is high. Therefore the production cost of MWCNTs material is reasonably lower than that of SWCNTs material [8]. Additionally, compared with SWCNTs, MWCNTs material has greater mechanical strength, better thermal stability and more photon absorption per nanotube due to higher mass density of the multi-walls. Therefore, the thermal or laser damage threshold of MWCNT should be higher than that of the SWCNTs.

However, there are only a few reported works on the application of MWCNTs material as a SA [7-8]. In this paper, a Q-switched Ytterbium doped fiber laser (YDFL) is demonstrated by using a new developed MWCNTs embedded in polyvinyl alcohol (PVA) film as a passive SA. The laser delivers Q-switched pulses with a repetition rate in a range from 5.484 kHz to 32.19 kHz with the maximum pulsed energy of 211.8 nJ.

2. Fabrication procedure and Raman characterisation of MWCNT-PVA film

The MWCNTs PVA SA used in our experiment is prepared as follows. First, the functionalizer solution is prepared by dissolving 4 g of sodium dodecyl sulphate (SDS) in 400 ml of deionized water. The diameter of the MWCNTs used is about 10–20 nm and the length is from 1 to 2 μm. 250 mg MWCNT is added to the functionalizer solution so that it can be dissolved in water. The homogenous dispersion of MWCNTs is achieved after the mixed solution is sonicated for 60 minutes at 50 W. The solution is then centrifuged at 1000 rpm to remove large particles of undispersed MWCNTs to obtain a stable dispersed suspension. Later, PVA solution is prepared by dissolving 1 g of PVA (Mw = 89×10^2 g/mol) in 120 ml of...
deionized water. MWCNTs-PVA composite is concocted by adding the dispersed MWCNTs suspension into the PVA solution by one to four ratio. After sonificating the mixture for more than one hour, a homogeneous MWCNTs-PVA composite was obtained. The MWCNT-PVA composite is casted onto a glass petri dish and left to dry at room temperature for about 24 hours to produce a thin film with a thickness around 50 μm. Fig. 1 shows the Raman spectrum of the MWCNTs film when it is excited by a 532 nm laser. The Raman spectrum of pure PVA film is also included in the figure for comparison purpose.

![Raman spectrum](image)

**Fig. 1. Raman spectroscopy of pure PVA and MWNTs-PVA composites film.**

As shown in Fig. 1, the Raman spectrum of the pure PVA film shows an intense peak at 2915 cm\(^{-1}\). For the fabricated MWCNTs-PVA film, the Raman spectrum shows the G and D bands at 1593 cm\(^{-1}\) and 1356 cm\(^{-1}\), respectively. The result is in good agreement to the values reported by Saito et. al. [9] and Kurti et al. [10]. The radial breathing mode (RBM), a distinctive feature of single-walled carbon nanotubes, is usually not observed in MWCNTs because the outer tubes restrict the breathing mode. The sample also produced a G’ peak at 2720 cm\(^{-1}\) which originates from two-phonon scattering phenomena in the Brillouin zone. For MWCNTs-PVA polymer composite, another intense peak is also observed at 2915 cm\(^{-1}\) due to the introduction of host polymer.

3. Experimental setup

Fig. 2 shows the configuration of the proposed Q-switched YDFL using a MWCNT/PVA composite film as SA. The MWCNT/PVA SA is fabricated by cutting a small piece of the prepared film (2×2 mm\(^2\)) and sandwiching it between two FC/PC fiber connectors, after depositing index-matching gel onto the fiber ends. The insertion loss of the SA is measured to be around 2 dB at 1050 nm. The laser employs a 2 m long Ytterbium-doped fiber (YDF) with a core diameter of 4.0 μm, NA of 0.20 and cutoff wavelength of around 980 nm as the gain medium. The doping level of ytterbium ions in the fiber is 1500 ppm. The YDF is pumped by a 980 nm laser diode through a 980/1050 nm wavelength division multiplexer (WDM). An isolator is used in the ring cavity to ensure unidirectional propagation of oscillating laser. The output of the laser is tapped out of the cavity through a 3 dB coupler. The optical spectrum analyser (OSA) with a spectral resolution of 0.02 nm is used for the spectral analysis of the Q-switched YDFL whereas the 350MHz oscilloscope is used to observe the output pulse train of the Q-switched operation via a 460-kHz bandwidth photodetector. The total cavity length of the ring resonator is around 12 m.
4. Results and discussion

The output spectrum of the passively Q-switched YDFL is investigated at various pump power and the result is shown in Fig. 3. From the figure, it can be seen that the laser operates at wavelength of 1058.6 nm as the pump power is increased above the threshold pump power of 65.72 mW. The peak power of the spectrum is observed to improve with the increase of pump power. The YDFL starts to generate a self-starting Q-switching pulse at threshold pump power of 65.72 mW. When the pump power is gradually increased from 65.72 to 97.29 mW, a stable pulse train with a varying repetition rate was observed. It is also observed that further increase of the pump power results in unstable pulsation. Figs. 4(a) and (b) shows the typical pulse train and its single pulse envelop of the laser output respectively, at pump power of 97.29 mW. The pulse shows the typical feature of passive Q-switching and no timing jitter is noticeable (limited by the sensitivity of 460-kHz detector). The pulse train has a period of 31.1 μs, which corresponds to a repetition rate of 32.19 kHz. The corresponding pulse envelop has the symmetrical Gaussian-like shape with full-width at half-maximum (FWHM) of 6.677 μs.

Fig. 2. Configuration of the proposed Q-switched YDFL utilizing a MWCNTs-PVA film based SA.

Fig. 3. Output spectra of the Q-switched YDFL at different pump powers.
Unlike passively mode-locking condition where the repetition rate of the output pulses is fixed corresponding to the cavity length [11], the repetition rate of a Q-switched fiber laser can be varied with reference to the lifetime of gain medium as well as pump power. Since different pump powers induce different time required to replenish the extracted energy between two consecutive pulses, the detuning of repetition rate occurs. Fig. 5 shows the repetition rate and average output power as a function of 980-nm pump power. It is found that both repetition rate and average output power monotonically increase with the pump power. The repetition rate is detuned from 5.48 to 32.19 kHz as the pump power increases from 65.72 to 97.29 mW, confirming the constructed laser is working under passive Q-switching. The output power increases from 1.05 mW to 5.64 mW as the pump power varies within the same range. This is attributed to the Ytterbium ions in the fiber, which are excited to a higher level even faster and the carbon nanotubes that are saturated faster as well which give a higher repetition rate.
Fig. 6 shows the pulse width and pulse energy of the Q-switched YDFL as a function of 980 nm pump power. As the pump power is increased from 65.72 mW to 69.71 mW, more gain is provided to saturate the SA, which in turn reduces the pulse width and increases the output pulse energy. However, the pulse energy fluctuates and then reduces with further increase in pump power. This is due to the pulse width of the output laser that exhibits increasing trend at this pump power region. As the pump power grows, the non-radiative decay rate of Yb$^{3+}$ ions also increases. This generates heat in the laser cavity where the MWCNTs end up absorbing. This will energize some electrons in the MWCNT’s valence band to move up to the conduction band from the strong internal thermal motion. As less electron in the valence band are available for photon absorption, the initial transmittance of the SA becomes greater as the pump power or absorbed heat increases. Thus MWCNT’s efficiency as SA drops such that the pulse width begins to increase as the pump power reaches 69.71 mW as shown in Fig. 6. The pulse width increases from 4.2 to 6.7 µs as the pump power is increased from 69.71 to 97.29 mW. The maximum pulse energy of 211.8 nJ is obtained at the pump power of 77.75 mW. The pulse energy could be improved by reducing the insertion loss of the SA or by optimizing the laser cavity. The Q-switching can be maintained up to a pump power of 97.29 mW, where the energy is larger than previously reported SWCNT based Q-switched fiber lasers [12]. The pulse becomes unstable and disappears as the pump power is further increased.

5. Conclusion

A passively Q-switched ring YDFL is demonstrated using MWCNTs embedded in PVA film as a saturable absorber. The SA film is prepared by mixing the MWCNTs homogeneous solution into a dilute PVA polymer solution. A small piece of the film is sandwiched between two FC/PC fiber connectors and integrated into the laser cavity to generate a stable Q-switching pulse at wavelength of 1058.6 nm within a pump power range of 65.72 to 97.29 mW. The repetition rate of the laser can be varied from 5.484 to 32.19 kHz by varying the pump power. The lowest pulse width of 4.2 µs is obtained at the pump power of 69.71 mW while the highest pulse energy of 211.8 nJ is obtained at pump power of 77.75 mW.

Acknowledgement

This work is financially supported by University of Malaya under various grant schemes (Grant No: RP008D-13AET and PG100-2014B) and Ministry of Science, Technology and Innovation (Grant No.: SF014-2014).

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Q-switched Ytterbium doped fiber laser using a multi-walled carbon nanotubes embedded in polyethylene oxide film


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Q-SWITCHED Yb-DOPED FIBER RING LASER WITH A SATURABLE ABSORBER BASED ON A GRAPHENE POLYVINYL ALCOHOL FILM

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Abstract

We propose, design, and demonstrate a Q-switched ytterbium-doped fiber laser (YDFL) employing a thin graphene polyvinyl alcohol (PVA) film as a passive saturable absorber (SA). The graphene is synthesized by electrochemical exfoliation of graphite at room temperature in 1% sodium dodecyl sulfate (SDS) aqueous solution. Graphene flakes obtained from the process are mixed with PVA solution as the host polymer to produce a thin film, which acts as a passive Q-switcher in the YDFL ring cavity. The laser generates a stable pulse operating at a wavelength of 1.076.4 nm with a threshold pump power of 73.7 mW. At a maximum 980 nm pump power of 113.6 mW, the YDFL generates an optical pulse train with a repetition rate of 25.53 kHz and a pulse width of 10 μs. The maximum pulse energy of 50.9 nJ is obtained at a pump power of 109.9 mW. A higher-performance Q-switched YDFL is expected to be achieved with optimization of the graphene saturable absorber and the laser cavity.

Keywords: graphene film, Q-switching, Yb-doped fiber laser.

1. Introduction

Development of Q-switched fiber lasers is an alternative to bulk pulsed solid-state lasers operating in the nanosecond to microsecond range for many applications, such as range finding, remote sensing, industrial processing, and medicine [1–3]. They can be achieved by numerous schemes based on active and passive approaches. Passively Q-switched fiber lasers feature a more compact geometry and simpler setup than active ones, which require additional switching electronics such as acousto-optic modulators [4]. It is typically achieved by inserting a saturable absorber (SA) into the cavity. Doped bulk crystals [5], semiconductor SA mirrors (SESAMs) [6], and carbon nanotubes (CNTs) [7] are the common SAs in passive Q-switching. However, doped crystals are mostly used in solid-state lasers. Doped crystals, such as SAs in fiber lasers, require extra elements (mirrors, lenses) to focus the fiber output into the crystal. Therefore, it is limited in a small range. SESAMs have limited operation bandwidth, typically a few tens nanometers [8], which are not suitable for broad-band tunable pulse generation. Compared to SESAMs,
CNTs can be considered as broadband SAs operated with Yb-doped [9] and Er-doped [10] fiber lasers. However, its operation wavelength is related to the diameter and chirality of the CNTs.

Recently, graphene, a single layer of carbon in a hexagonal lattice, has been intensely studied due to its wonderful optical properties [11]. In laser photonics, graphene SA are widely used as a broadband SA for passively $Q$-switching or mode locking the fiber lasers or solid state lasers at different laser wavelengths ranging from 1 to 2 $\mu$m [12,13]. To date, many works have been reported on the integration of a graphene polyvinyl alcohol (PVA) SA into the fiber-laser system for ultra-short-pulse generation. For instance, a mode-locked erbium-doped fiber laser with a stable soliton-like pulse was achieved using graphene from chemical vapor deposition (CVD) process [14].

In this paper, we design and demonstrate for the first time a $Q$-switched ytterbium-doped fiber laser employing a graphene PVA-based saturable absorber. The saturable absorber is based on a graphene film, which is fabricated from graphene flakes obtained from electrochemical exfoliation. The SA is integrated in the YDFL by sandwiching the graphene thin film between two fiber connectors to achieve a stable pulse train operating at 1,076.4 nm with a threshold pump power of 73.7 mW. The laser generates $Q$-switched pulses with 25.53 kHz repetition rate, 10 $\mu$s pulse width, and 46.9 nJ pulse energy employing a pump power of 113.6 mW at 980 nm.

2. Experimental Procedure

In this work, the key part of $Q$-switched generation is the fabrication of a saturable absorber film based on a graphene sheet in the polyvinyl alcohol (PVA) host. The PVA is a water-soluble synthetic polymer with monomer formula $C_2H_4O$, which has excellent film forming, emulsifying, and adhesive properties. It also has high tensile strength, flexibility, and high oxygen and aroma barrier, although these properties depend on the humidity.

First, graphene flakes were produced using the electrochemical exfoliation process. In this process, a constant voltage difference of 20 V is applied to two electrodes (graphite rods), which are placed 1 cm apart in an electrolysis cell filled with electrolyte (1% SDS in deionized water). Hydroxyl and oxygen radicals are generated due to electrolysis of the water at the electrode during the electrochemical process. Then the oxygen radicals start to corrode the graphite anode, and this is followed by the intercalation of anionic surfactant. Finally, graphene sheets are created in the solution.

In our work, black sediments (graphene) start to peel off from the anode after several minutes. The exfoliation process is continued for 2 h for obtaining a stable graphene suspension in the SDS solution. The stable graphene suspension is centrifuged at 3,000 rpm for 30 min to remove large agglomerates. Afterwards, the supernatant portion of the suspension is decanted. Second, we prepare a PVA solution by dissolving 1 g of PVA in 120 ml distilled water. Then we mix the PVA solution with the centrifuged graphene suspension at a ratio of 3:2 (in ml) to form the precursor. Then the mixture is stirred using an ultrasonic cleaner for about 1 h. This step helps to get precursor with enough viscosity so that it can be easily used in forming a film. Finally, suitable amounts of precursor are spread thinly on the glass substrate and allowed to dry in an oven with temperature of 56$^\circ$ to form the graphene PVA film, which is then used to construct a saturable absorber. The thickness of the film is $\sim 50$ $\mu$m.

The experimental setup of the proposed $Q$-switched YDFL is shown in Fig. 1; it consists of a 7 m long ytterbium-doped fiber (YDF), a wavelength division multiplexer (WDM), a polarization controller, an isolator, a graphene PVA film based SA, and 3 dB output coupler in a ring configuration. The SA is fabricated by cutting a small part of the earlier prepared film (2x2 mm$^2$) and sandwiching it between two
FC/PC fiber connectors, after depositing refractive index matching gel onto the fiber ends. The insertion losses of the SA are measured to be around 1.5 dB at 1,100 nm. The YDF used has a core and cladding diameters of 4 and 125 μm, respectively, a numerical aperture of 0.20, and ytterbium ion absorption of 280 dB/m at 920 nm. It is pumped by a 980 nm laser diode via the WDM. An isolator is incorporated in the laser cavity to ensure unidirectional propagation of the oscillating laser. A PC is used to optimize Q-switching and control the laser stability. The laser output is tapped out of the cavity through a 3 dB coupler. The optical spectral analyzer is used to analyze the spectra of the Q-switched YDFL with a spectral resolution of 0.02 nm, whereas the oscilloscope is used to observe the output pulse train of the Q-switched operation via a 460 kHz bandwidth photodetector (Thor lab, PDA50B-EC). The total cavity length of the ring resonator is measured to be around 14 m.

3. Results and Discussion

Raman spectroscopy was performed to confirm the presence of the graphene layer in the fabricated graphene PVA film. In Fig. 2, we show the obtained Raman spectrum, which clearly indicates the three prominent peaks. These peaks are located at approximately 1,341, 1,603, and 2,910 cm\(^{-1}\), generally known as the D, G, and 2D band, respectively. The G band contributes to the \(E_{2g}\) mode of graphite and is related to the in-plane vibration of sp\(^2\)-bonded carbon atoms, while the D band is associated with the vibrations of carbon atoms with the sp\(^3\) electronic configuration of disordered graphite. The intensity ratio of the D and G bands of the graphene sheets is about 1.2, indicating defects of the graphene samples. However, the amount of structural defects is not large, since the D peak is not very broad [15]. The intensity ratio between the G and D peaks can be used to determine the number of graphene layer. It was reported that the single-layer graphene had a low intensity ratio, usually lower than 0.5, while multilayer graphene shows a higher intensity ratio (≥ 1) [16]. As indicated in Fig. 2, we obtained a G/2D peak ratio of 0.6, which indicates a multilayer structure of the graphene. The shape of the 2D peak can also be used to estimate the number of graphene layers. As the graphene layer increases, the full-width at half maximum (FWHM) of the 2D peak follows [17]. As also shown in Fig. 2, the FWHM for the G and 2D peaks are obtained at 50 and 104 cm\(^{-1}\), respectively.
Stable and self-starting Q-switching operation is obtained just by adjusting the pump power over a threshold of 73.7 mW. In Fig. 3 (left), we show the output spectrum of the Q-switched laser at a pump power of 105 mW. As seen, the laser operates at a wavelength of 1,076.4 nm with the optical signal-to-noise ratio (OSNR) greater than 40 dB. In Fig. 3 (right), we show the average output power of the laser against the pump power. The output power increases from 0.22 to 1.15 mW as the pump power increases from 73.7 to 113.6 mW. The pump power was limited to 113.6 mW to avoid damage to the graphene SA due to the thermal effect [18].

In Fig. 4, we show oscilloscope traces of the Q-switched pulse train at a pump power of 73.7 mW. There is no distinct amplitude modulation in each envelop of the Q-switched spectrum, which means that the self-mode locking effect on the Q-switching is weak. At the threshold pump power, a stable passively Q-switching operation starts to occur with an average output power of 0.22 mW, a pulse width of 14.38 μs, and a repetition rate of 10.28 kHz. The pulse energy is calculated to be around 21.7 nJ at this pump power. At the maximum 980 nm pump power of 113.6 mW, the Q-switched laser produces a pulse train with the highest repetition rate of 25.53 kHz and the shortest pulse width of around 10.0 μs.

In Fig. 5, we show the envelop of the Q-switched pulse at the maximum pump power of 113.6 mW.

In Fig. 6, we show the repetition rate and the pulse width versus the pump power. The repetition...
Fig. 6. Repetition rate and pulse width as a function of the pump power at a wavelength of 1,076.4 nm.

Fig. 7. Pulse energy as a function of the pump power.

rate of the graphene-based Q-switched YDFL is monotonically increasing, demonstrating a near linear dependence on the pump power level, which is consistent with the result of EDFL for 1,550 nm operation [19]. When the pump power is tuned from 73.7 to 113.6 mW, the pulse-train repetition rate varies from 10.3 to 24.5 kHz. On the other hand, the pulse width is inversely proportional to the pump power, where the pulse duration becomes shorter as the pump power increases. The pulse width monotonically decreases from 14.1 to 10.0 μs as the pump power increases from the threshold value to the maximum pump power of 113.6 mW. The pulse width is expected to decrease further if the pump power can be augmented beyond 113.6 mW as long as it is still kept below the damage threshold of the graphene-based SA. The duration of the long pulse is mainly relative to the long length of the cavity, which resulted in a relatively longer cavity lifetime [20]. Shortening the total cavity length of the fiber laser is another alternative to getting a shorter pulse.

In Fig. 7, we show the pulse energy against the pump power. As seen, the pulse energy demonstrates an increasing trend with increase in the pump power. The maximum pulse energy of 50.9 nJ was obtained at a pump power of 109.9 mW. The pulse energy could be improved by reducing the insertion losses of the graphene saturable absorber and optimizing the laser cavity.

4. Conclusions

We demonstrated experimentally a Q-switched Yb-doped fiber laser operating at 1,076.4 nm using a graphene PVA film based saturable absorber. The graphene suspension was produced by an electrochemical exfoliation process and mixed with PVA solution as the host polymer to produce a thin film, which acts as a passive Q-switcher in the YDFL ring cavity. When the pump power is tuned from a threshold pump power of 73.7 mW to a value of 113.6 mW, the pulse-train repetition rate varies from 10.3 to 24.5 kHz. On the other hand, the pulse width monotonically decreases from 14.1 to 10.0 μs as the pump power increases from the threshold value to the maximum pump power of 113.6 mW. We obtained a maximum pulse energy of 50.9 nJ at a pump power of 109.9 mW.

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Q-Switched Ytterbium-Doped Fiber Laser Using Black Phosphorus as Saturable Absorber

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Q-Switched Ytterbium-Doped Fiber Laser Using Black Phosphorus as Saturable Absorber

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(Received 29 January 2016)

We demonstrate a Q-switched ytterbium-doped fiber laser (YDFL) using a newly developed multi-layer black phosphorous (BP) saturable absorber (SA). The BP SA is prepared by mechanically exfoliating a BP crystal and sticking the acquired BP flakes onto a scotch tape. A small piece of the tape is then placed between two ferrules and incorporated in a YDFL cavity to achieve a stable Q-switched operation in a 1.0 µm region. The laser has a pump threshold of 55.1 mW, a pulse repetition rate that is tunable from 8.2 to 32.9 kHz, and the narrowest pulse width of 10.8 µs. The highest pulse energy of 328 nJ is achieved at the pump power of 97.6 mW. Our results show that multi-layer BP is a promising SA for Q-switching laser operation.

PACS: 42.60.Fc, 42.65.Re, 42.65.Tg DOI: 10.1088/0256-307X/33/5/054206

Passively Q-switched fiber lasers have gained widespread attention due to their advantages in compactness and flexibility, and potential applications in medicine, telecommunications, remote sensing and laser material processing. To date, many forms of saturable absorber (SA) have been used in generating passive Q-switching including semiconductor saturable absorber mirror (SESAM),[1] single-walled carbon nanotubes (SWCNTs)[2] and graphene.[3,4] Despite the successful implementation of these SAs, researchers are still looking for new SAs with better characteristics in terms of wavelength-independent saturable absorption, low saturable optical intensity, fiber compatibility and high damage threshold. Recently, topological insulators (TIs) and molybdenum disulfide (MoS2) have also been identified as potential SAs for mode-locking and Q-switching.[5–7]

More recently, black phosphorus (BP), dubbed as the newly emerging allotrope, has gained much attention for potential applications in the next-generation optoelectronic devices such as sensors, field-effect transistors and solar cells.[8–10] Multi-layer BP has a similar structure as bulk graphite. In a single layer, each phosphorus atom is covalently bonded with three adjacent phosphorus atoms to form a stable honeycomb structure, and different layers are stacked together by van der Waals interaction.[11] It has the common properties of 2D materials such as wideband absorption, ultrafast carrier dynamics and planar characteristic.[12] The BP comprises only the elemental ‘phosphorus’ and thus it could be easily peeled off by mechanical exfoliation. In this Letter, we report a passively Q-switched ytterbium-doped fiber (YDF) laser generated using multi-layer BP-based SA, obtained by the mechanical exfoliation method. The SA is prepared by peeling bulk-structured BP 2D thin layers using a scotch tape. By inserting a small piece of the BP-SA tape into the ytterbium-doped fiber laser cavity, Q-switched pulses with a maximum pulse energy of 328 nJ are achieved, with the shortest pulse width of 10.8 µs. Mechanical exfoliation is advantageous mainly due to its simplicity and reliability, where the entire fabrication process is free from complicated chemical procedures and costly instruments.

The multi-layer BP used in our experiment was prepared by the mechanical exfoliation technique in the following. Initially, we peeled off thin flakes from a large block of commercially available BP crystal (purity of 99.995%) using the clear scotch tape. Then, we repeatedly pressed the flakes on the scotch tape to form a thin layer of BP, thin enough to transmit light with a high efficiency. Afterwards, a small piece of the BP tape was cut out and attached onto a standard FC/PC fiber ferrule end surface with the aid of index matching gel. After connecting it with another FC/PC fiber ferrule with a standard flange adapter, the all-fiber BP-based SA was finally ready. The entire preparation process is illustrated in Fig. 1.

The composition of the multi-layer BP tape is examined by the energy dispersive spectroscopy (EDS) on the field emission scanning electron microscopy (FESEM). Presence of the BP material on the scotch tape adhesive surface was confirmed by the presence of the high peak of phosphorus in the spectroscopy as shown in Fig. 2. The inset in Fig. 2 shows the FESEM image, which confirms the existence of uniform multi-layer phosphorus and the absence of >1 µm aggregates or voids in the tape, which otherwise result in non-saturable scattering losses. We also performed Raman spectroscopy on the prepared BP tape sample. Figure 3 shows the Raman spectrum, which is recorded by a spectrometer when a 514 nm beam of an argon ion laser is directed on the tape for 10 ms with an exposure power of 10 mW. As shown in the figure, the sample exhibits three distinct Raman peaks at 359 cm⁻¹, 434 cm⁻¹ and 460 cm⁻¹,[13] correspond-
ing to $B_{2g}$ and vibration modes of a layered BP. While $B_{2g}$ and modes correspond to the in-plane oscillation of phosphorus atoms in the BP layer, the mode corresponds to the out-of-plane vibration. The ratio between peak and silicon (Si) level at 520 cm$^{-1}$ indicates that the thickness of the BP flake is $\sim$200 nm. The thickness for single-layer BP is about 0.8 nm; thus the prepared BP-based SA is expected to consist of 250 layers.

![Image](a) The preparation flow of BP-SA by using the mechanical exfoliation method. (a) The peeled BP crystal is placed onto a sticky tape. (b) The crystal is thinned to the small flakes by repeatedly pressing the flakes onto a sticky tape. (c) The image of the BP tape after the thinning process. (d) Attaching the BP tape onto the ferrule.

![Image](b) EDS from the FESEM image of the prepared BP tape, which confirms the presence of phosphorus. Inset: the FESEM image.

![Image](c) Raman spectrum of the multi-layered BP tape.

The nonlinear optical response property for the multi-layer BP on the scotch tape was also investigated to confirm its saturable absorption by applying an absorption technique. A self-contracted mode-locked fiber laser (1557 nm wavelength, 1.5 ps pulsewidth, 17.4 MHz repetition rate) was used as the input pulse source. The transmitted power and a reference power for normalization are recorded as a function of incident intensity on the tape by varying the input laser power. With increasing the peak intensity, the material absorption decreases as shown in Fig. 4, confirming the saturable absorption characteristic of the multi-layer BP. The experimental results for absorption are fitted according to a simple two-level SA model [13]

$$\alpha(I) = \frac{\alpha_s}{1 + I/I_{sat}} + \alpha_{ns},$$

where $\alpha(I)$ is the absorption, $\alpha_s$ is the modulation depth, $I$ is the input intensity, $I_{sat}$ is the saturation intensity, and $\alpha_{ns}$ is the non-saturable absorption. As shown in Fig. 4, the modulation depth, non-saturable intensity, and saturation intensity are 7%, 58% and 0.0025 MW/cm$^2$, respectively. Taking into account its nonlinear optical response leading to absorption saturation at relatively low intensity, the mechanically exfoliated BP meets basic criteria of a passive SA for fiber lasers.

![Image](d) Nonlinear saturable absorption profile of the multi-layer BP.

The experimental setup of the proposed Q-switched YDF fiber ring laser using SA.

The experimental setup of the proposed Q-switched YDF laser is schematically shown in Fig. 5. The ring cavity consists of a 1.5-m-long YDF, a wavelength division multiplexer (WDM), an isolator, a 3dB output coupler and the multi-layer BP Q-switcher. The total cavity length is about 6 m. The YDF used has a core diameter of 4.0 $\mu$m, NA of 0.20 and cutoff wavelength of around 980 nm. The doping level of ytterbium ions in the fiber is 1500 ppm. The YDF is pumped by a 980 nm pump laser diode through a 980/1030 nm WDM. The multi-layer BP on the scotch tape is integrated into the cavity as a passive Q-switcher while a polarization insensitve optical isolator is incorporated to ensure unidirectional operation of the laser. The Q-switched pulse train is taken out via a 3dB output coupler which keeps half the light oscillating in the ring cavity for both spectral and temporal diagnostics. The output is simultaneously monitored by using a 350-MHz oscil-
The YDF laser started to operate in the cw mode at the pump power of 40.5 mW, before transitioning to the Q-switching operation at 55.1 mW. When the pump power was gradually increased up to 97.6 mW, a stable pulse train with an increasing repetition rate was observed. This is a typical feature of passive Q-switching. Figure 6 shows the spectral and temporal characteristics of the output Q-switching pulse train when the 980 nm pump power was fixed at 76.6 mW. The output spectrum is depicted in Fig. 6(a), which indicates that the Q-switched laser operates at 1069.4 nm with 3 dB spectral bandwidth of about 1.0 nm and the signal-to-noise ratio of about 40 dB. Figure 6(b) shows the typical oscilloscope trace of the Q-switched pulses. The pulse train has the period of 38.3 μs without noticeable timing jitter. Figure 6(c) reveals a typical pulse envelope, displaying an almost symmetrical Gaussian-like shape with a pulse duration of 12.2 μs. The rf output spectrum in Fig. 6(d) shows that the Q-switched pulses have a repetition rate of 26.1 kHz, which agrees with the pulse period of 38.3 μs measured in Fig. 6(b). It is also observed that the rf signal-to-noise ratio is about 23 dB with no spectral modulation, indicating that the passively Q-switching operation was stable.

Figure 7 shows the recorded pulse repetition rate and the pulse duration as a function of the pump power. The repetition rate of the Q-switching pulses linearly increases from 8.1 to 32.9 kHz as the pump power is increased from 55.1 to 82.0 mW. However, the repetition rate reduces as the pump power is further increased before the pulse train becomes unstable, and disappears as the pump power is increased above 97.6 mW. Meanwhile, the pulse duration significantly decreases from 17.9 to 10.8 μs as the pump power is increased from 55.1 to 97.6 mW. The pulse duration could be further narrowed by optimizing the parameters such as shortening the cavity length and improving the modulation depth of the BP Q-switcher. It is observed in Fig. 8 that both average output power and pulse energy almost linearly increase with the input pump power up to the pump power of 97.6 mW. The maximum average output power and pulse energy are 10 mW and 328 nJ, respectively. The experiment is also repeated for different locations of the BP tape sample. It is found that a small deviation of tape or BP layer thickness does not change the optical properties of the BP SA and the Q-switching performance. We believe that the pulse energy produced by the laser could be further improved by optimizing the SA parameters of BP tape and the cavity design. The output pulse energy could be further improved if the cavity loss and SA loss can be further improved. The similar performance of Q-switching is also obtained in the erbium-doped fiber laser (EDFL) and the thulium-doped fiber laser (TDFL) setups, which operate at 1550 nm and 1920 nm, respectively. This shows that the proposed BP SA can be used as a wideband SA.

In conclusion, we have experimentally demonstrated that a multi-layer BP tape can be used as an effective SA for passive Q-switching. The BP tape is prepared by the mechanical exfoliation method and sandwiched between two fiber ferrules via a fiber adapter to form a fiber-compatible SA. Stable Q-switched pulses at 1069.4 nm are successfully obtained. The laser registers a pump threshold of 55.1 mW and a minimum pulse duration of 10.8 μs. The pulse repetition rate could be varied over a wide range of frequencies, from 8.2 to 32.9 kHz, by adjusting the pump power. The highest pulse energy of 328 nJ is obtained at the pump power of 97.6 mW. Our ex-
Experimental results suggest that a multi-layer BP is a promising material for pulsed laser applications.

References

Passively Q-switched Ytterbium doped fiber laser with mechanically exfoliated MoS$_2$ saturable absorber

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Passively Q-switched Ytterbium doped fiber laser with mechanically exfoliated MoS$_2$ saturable absorber

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Received: 10 July 2016 / Accepted: 10 November 2016 / Published online: 9 January 2017

Abstract: A passively Q-switched Ytterbium-doped fiber laser (YDFL) based on MoS$_2$ saturable absorber (SA) is demonstrated. A few layers of MoS$_2$ are mechanically exfoliated from a natural MoS$_2$ crystal using a scotch tape and the resulting SA is sandwiched between two fiber ferrules to form a fiber compatible Q-switcher. The saturation intensity, non-saturable intensity, and modulation depth of the MoS$_2$ SA are 23.5 MW/cm$^2$, 23.0, and 11.3%, respectively. By introducing the MoS$_2$ SA into the YDFL cavity, a stable pulse laser is generated at 1070.2 nm wavelength with a threshold pump power of 49.57 mW. The repetition rate of the Q-switched pulses ranges from 3.817 to 25.25 kHz, as the 980 nm pump power increases from 49.57 to 87.2 mW. The highest pulse energy is 295.45 nJ at a pump power of 87.2 mW.

Keywords: Molybdenum disulfide; Layers MoS$_2$; Passive Q-switcher

PACS Nos.: 42.81.-I; 42.60.Gd; 42.60.Da; 42.55.Wd

1. Introduction

Compact and flexible passively Q-switched fiber lasers have opened up new applications in laser material processing, remote sensing, telecommunications and medicine [1–3]. One of the most effective methods for generating passive Q-switching in fiber lasers is by using a saturable absorber (SA). In the past two decades, various SAs namely semiconductor saturable absorber mirror SESAM [4], carbon nanotubes CNTs [5, 6] and graphene [7–9], have been successfully used to passively generate Q-switched fiber lasers. Even today, researchers are still searching for new SAs which possess desirable characteristics such as wavelength-independent saturable absorption, low saturable optical intensity, high damage threshold, and excellent fiber compatibility.

More recently, Molybdenum Disulfide (MoS$_2$) has also been tested as an SA for mode-locking and Q-switching [10–17]. Wang et al. [10] have investigated the saturable absorption property of two-dimensional (2D) MoS$_2$ nanosheets for ultrafast laser applications. Other reported works indicate that MoS$_2$ has a higher modulation depth than that of graphene ($\sim$2.3% per layer) [18–21]. Zhang et al. [11] have demonstrated a mode-locked Ytterbium-doped fiber laser (YDFL) that emitted dissipative solitons at 1.054 $\mu$m using a MoS$_2$ SA. Liu et al. [12] have achieved femtosecond pulses from a mode-locked EDFL with a MoS$_2$-polymer composite SA. For 1 $\mu$m region Q-switched fiber lasers using MoS$_2$ as SAs, so far only a maximum pulse energy of 126 nJ is reported [13, 15]. In this paper, we describe a passive Q-switched YDFL operating at 1070.2 nm with a high pulse energy of 295.45 nJ. The Q-switched laser performs competitively compared to similar lasers in 1 $\mu$m region that use other 2D materials as SAs [21, 22].

2. Preparation of MoS$_2$ SA and its optical characteristic

MoS$_2$ is a nanomaterial which has extraordinary electronic and optical properties [23]. The MoS$_2$ layers used in the construction of the SA were obtained by mechanical exfoliation method [24]. The fabrication process was performed without using any chemical substance and...
costly instrument. Fig. 1(a) shows the relatively thin flakes that is evenly peeled off a natural MoS$_2$ crystal by scotch tape. The flakes attached to the scotch tape was repeatedly pressed to make it thin enough to transmit light with a high efficiency. Then a tiny piece of the MoS$_2$ tape was cut and placed onto the end face of a FC/PC fiber connector ferrule after applying index matching gel as shown in Fig. 1(b). After connecting it with another FC/PC ferrule with a standard flange adapter, the all-fiber MoS$_2$ SA was finally completed and ready to be used. It is learned that if a clean scotch tape is placed onto the end face of fiber connector, the cavity will fail to generate Q-switching regime.

Raman spectroscopy was performed on the fabricated MoS$_2$ SA. Fig. 2(a) shows the Raman spectrum recorded by a spectrometer when a 514 nm beam of an Argon laser was radiated on the MoS$_2$ tape for 10 s with an exposure power of 10 mW. As shown in the figure, the sample exhibits two characteristic peaks in parallel with two phonon modes; out of plane vibration of Sulfide atoms at 408 cm$^{-1}$, and in plane vibration of Molybdenum and Sulfide atoms at 383 cm$^{-1}$ with a frequency difference of 25 cm$^{-1}$. This frequency difference indicates that the prepared tape contains the layers of MoS$_2$ [25]. Following other reported results [26], the full-width-half-maximum (FWHM) of $E_{2g}$ and $A_{1g}$ bands were calculated to be 5.0

Fig. 1 Mechanical exfoliation method; (a) simple peeling process, and (b) MoS$_2$ tape at standard FC/PC fiber end surface

(a)

(b)
and 5.5 cm\(^{-1}\), respectively. As shown in Fig. 2(b), the linear absorption of the SA ranges from 1520 to 1600 nm. The figure also shows that the absorption looks rather linear as it follows alat profile. The inset of Fig. 2(b) shows the absorption spectra within UV–Vis region. From the figure, two exciton peaks of MoS\(_2\) are noticeable at 612 and 670 nm. These peaks correspond to the interband excitonics transition at 2.03 and 1.85 eV, respectively. The absorption spectra obtained are comparable to those found in previous studies.

The nonlinear optical response property of the MoS\(_2\)-SA tape was further investigated to confirm its saturable absorption. This was done by applying a balanced twin-detector measurement technique. A self-constructed mode-locked fiber laser (1557 nm wavelength, 1.5 ps pulsewidth, 17.4 MHz repetition rate) was used as the input pulse source. As in the work of Luo et al. [15], a 1566 nm mode-locked laser source was launched to generate lasers at 1, 1.5 and 2 \(\mu\)m. In our work, the transmitted power was recorded as a function of incident intensity on the device by varying the input laser power. The experimental data for transmission were fitted to a simple two-level SA model of

\[
T(I) = \frac{1}{C_0} a_{ns} \exp(-I/C_0) + \frac{1}{C_0} a_{ns} \exp(-I/C_0 - I_{sat})
\]

where \(T(I)\) is the transmission, \(a_{ns}\) is the modulation depth, \(I\) is the input intensity, \(I_{sat}\) is the saturation intensity, and \(a_{ns}\) is the non-saturable absorption. The nonlinear transmission of the MoS\(_2\) on the scotch tape is shown in Fig. 2(c). As seen, the modulation depth, non-saturable intensity, and saturation intensity are 11.3, 23.0%, and 23.5 MW/cm\(^2\), respectively. The large modulation depth of 11.3% is expected to suppress wave breaking in the mode-locked fiber laser and thus improves the attainable pulse energy.
3. Experimental setup

The experimental setup of the proposed MoS$_2$ based Q-switched YDFL is schematically shown in Fig. 3. The laser cavity consists of a 980/1030 nm wavelength division multiplexer (WDM), 1.5-m long of Ytterbium-doped fiber (YDF), MoS$_2$ SA, an isolator and a 3 dB output coupler in a ring configuration. The YDF used as a gain medium, has a numerical aperture of 0.20, a core and cladding diameters of 4 and 125 µm, respectively, as well as ion absorption of 280 dB/m at 920 nm. A unidirectional operation of the laser cavity is obtained by using an isolator. The output of the YDFL is collected from the cavity via a 50:50 coupler, which retains 50% of the light in the laser ring cavity for oscillation. The optical spectrum analyser (OSA) used for analyzing the Q-switching has a spectral resolution of
0.02 nm. The oscilloscope is used to observe the output pulse train of the Q-switched laser.

4. Results and discussion

The YDFL started lasing in Q-switched mode at a pump power of 49.57 mW. When the pump power is gradually increased from 49.57 to 87.2 mW, the repetition rate of the pulse increased in response. This is a typical feature of passive Q-switching. Fig. 4 shows the optical spectra of the Q-switched YDFL at three different pump powers of 49.57, 71.3, and 87.2 mW. While the peak of the YDFL stays fixed at 1070.2 nm, its 3 dB spectral bandwidth widens with the increase in pump power. The bandwidth are 0.2, 0.4 and 0.5 nm at the three pump powers respectively. To verify that the passive Q-switching is attributed to the MoS\textsubscript{2} SA, the MoS\textsubscript{2} tape is removed from the ring cavity. In this case, no Q-switched pulses are observed on the oscilloscope, even when the pump power is adjusted to a wide range. This finding confirms that the MoS\textsubscript{2} SA is responsible for the passively Q-switched operation of the laser.

Figures 5(a)–5(c) show the pulse trains and single-pulse envelops of the Q-switched YDFL at pump powers of 49.57, 71.3, and 87.2 mW, respectively. As the pump power increases, more energy can be stored in the laser cavity and this contributes to the rise in the repetition rate accompanied by the reduction in pulse width [27]. For pump powers of 49.57, 71.3, and 87.2 mW, the attained repetition rates are 3.817–17.54 and 25.25 kHz while the corresponding pulse widths are 26.09, 11.27 and 11.33 \(\mu\)s, respectively. The attributes of the pulse a Q-switched laser mainly depend on the gain medium, SA and pump power.

Figure 6 shows the repetition rate and pulse width of the Q-switched YDFL as a function of the pump power. As the pump power increases from 49.57 to 87.2 mW, the repetition rate grows from 3.817 to 25.25 kHz. Throughout the experiment, the output is stable and no amplitude modulations are observed. This indicates that there is no self-mode locking (SML) effect during the Q-switching operation. On the other hand, the pulse width decreases from 26.09 to 11.33 \(\mu\)s. As the pump power exceeds 71.3 mW, the pulse width stays nearly unchanged, which clearly indicates that the SA is saturated. It is reported that the pulse width of a passively Q-switched laser depends on the cavity round-trip time and the modulation depth of the SA [28]. Therefore, the minimum pulse duration obtained in our experiment can surely be narrowed by shortening the cavity length and improving the modulation depth of the MoS\textsubscript{2} layer.

Figure 7 shows the dependency between average output power and single pulse energy of the proposed Q-switched YDFL against the pump power. As shown, the average output power increases linearly from 0.542 to 7.46 mW while the pulse energy also increases from 142 to 295.45 nJ as the pump power rises from 49.57 to 87.2 mW. The pulse energy is calculated based on the measured average output power and the repetition rate. When the pump power goes past 87.2 mW, the Q-switched pulse train becomes unstable or disappears, as typically observed in certain passively Q-switched fiber lasers [3]. Subsequently, when the pump power is reduced to a level below...
87.2 mW, the stable Q-switched pulse train reappear. Hence, the unstable Q-switched operation at high pump power may be attributed to over-saturation of the MoS$_2$ SA rather than to thermal damage [3]. Fig. 8 shows the RF spectrum of the Q-switched YDFL at the pump power of 87.2 mW, for which the pulse repetition rate is 25.25 kHz, matching the oscilloscope data of Fig. 5(c). The RF signal-to-noise ratio (SNR) is about 30 dB with no spectral modulation, indicating that the passively Q-switching operation is stable.

5. Conclusion

We proposed and experimentally demonstrated a Q-switched YDFL operating at 1 μm region using a MoS$_2$ based SA. The bulk layer MoS$_2$ tape was fabricated by mechanical exfoliation technique and then sandwiched between two fiber ferrules to form the fiber compatible MoS$_2$ based Q-switcher. A Q-switched pulse train operating at 1070.2 nm was obtained as the 980 nm pump power reached the threshold value of 49.57 mW and it continued to operate stably until 87.2 mW. The corresponding pulse repetition rate was tunable from 3.817 to 25.25 kHz. The lowest pulse width was 11.33 μs and the maximum pulse energy was 295.45 nJ. Our experimental results suggest that MoS$_2$ is a promising material for pulsed laser applications.

Acknowledgements This work is financially supported by University of Malaya under various grant schemes (Grant Nos. RP008D-13AET and PG100-2014B).

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Q-switched ytterbium-doped fiber laser with topological insulator-based saturable absorber

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Q-switched ytterbium-doped fiber laser with topological insulator-based saturable absorber

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Abstract. A stable passive Q-switched ytterbium-doped fiber laser (YDFL) operating at 1057.4 and 1044.5 nm is demonstrated using two types of topological insulator-based saturable absorbers (SAs): bismuth (III) selenide ($\text{Bi}_2\text{Se}_3$) and bismuth (III) telluride ($\text{Bi}_2\text{Te}_3$). These materials are embedded in polyvinyl alcohol film and incorporated in the YDFL cavity. Using $\text{Bi}_2\text{Se}_3$ SA, the YDFL obtains a stable train of pulses with a repetition rate that is tunable from 48.83 to 102 kHz. Meanwhile, the corresponding pulse width decreases from 8.9 to 3.48 $\mu$s as the pump power increases from 67.6 to 88.3 mW. For the $\text{Bi}_2\text{Te}_3$-based Q-switched YDFL, the repetition rate grows from 43.25 to 93.63 kHz and the pulse width shrinks from 5.65 to 2.01 $\mu$s as the 980-nm pump power rises from 72.8 to 98.4 mW. The maximum pulse energy is obtained with the $\text{Bi}_2\text{Te}_3$ SA at 29.95 nJ. © 2017 Society of Photo-Optical Instrumentation Engineers (SPIE) [DOI: 10.1117/1.OE.56.5.056103]

Keywords: ytterbium-doped fiber laser; Q-switching; topological insulator; bismuth (III) selenide saturable absorber; bismuth (III) telluride.

1 Introduction

In recent years, significant research efforts have been devoted to the generation of short pulse fiber lasers by using various types of gain media and saturable absorbers (SA).1,2 For instance, Q-switched fiber lasers have been demonstrated using ytterbium-, erbium- and thulium-doped fiber as gain media to operate at 1-, 1.5-, and 2-$\mu$m region, respectively.3–5 The use of passive SAs, such as semiconductor SA mirrors (SESAMs),6 carbon nanotubes (CNTs),7,8 and graphene,9,10 has attracted much attention due to their considerable saturable absorption rate, ultrafast timing recovery, and a good compatibility with other components in the setup. SESAMs are well known for their stability and flexibility. However, fabricating an SESAM is complex and it requires expensive facilities. Furthermore, the operating range of an SESAM or its optical response is usually limited. Thus, CNTs and graphene-based SAs are preferable since they are cheaper and easier to fabricate.11,12

More recently, passive SAs made of topological insulators (TIs) have been shown to be capable of generating Q-switched and mode-locked lasers.13,14 Since TIs have a large modulation depth with an efficient saturable absorption property, they are suitable for making SAs. For instance, bismuth (III) telluride ($\text{Bi}_2\text{Te}_3$) has the modulation depth and saturation intensity of about 22% and 57 MW/cm², respectively.15 Previously, Q-switched fiber lasers were demonstrated based on erbium15,16 and ytterbium-doped fiber (YDF) using bismuth (III) selenide ($\text{Bi}_2\text{Se}_3$)16 as SA. The Q-switched YDF laser produces a pulse train with the repetition rate in a range from 8.3 to 29.1 kHz and maximum pulse energy of 17.9 nJ.

In this paper, two different passively Q-switched ytterbium-doped fiber laser (YDFL) are demonstrated using $\text{Bi}_2\text{Se}_3$ and $\text{Bi}_2\text{Te}_3$ nanoparticles-based SAs to exploit the wideband saturable absorption characteristic of the TIs. For this purpose, we prepare free-standing $\text{Bi}_2\text{Se}_3$- and $\text{Bi}_2\text{Te}_3$-polymer composite SA films by implanting the powder of these materials into a polyvinyl alcohol (PVA) host. These films are used in a fully fiber-integrated laser cavity for generating Q-switching pulses train. Compared with the previous YDFL,16 we obtained a higher repetition rate in both fiber lasers.

2 Preparation and Optical Characterization of Saturable Absorbers

The commercially available TI powders of $\text{Bi}_2\text{Se}_3$ and $\text{Bi}_2\text{Te}_3$ with a molecular weight of 654.84 and 800.76 g/mol, respectively, are used to prepare the SA in our experiment. To prepare the host polymer, 1 g of PVA (Sigma-Aldrich) is dissolved in 120 ml deionized water with the aid of a magnetic stirrer at room temperature. Next, 14 mg of $\text{Bi}_2\text{Se}_3$ powder and 14 mg of $\text{Bi}_2\text{Te}_3$ powder are mixed with 3 ml of the PVA solution separately. They are thoroughly mixed using a magnetic stirrer for 3 h. Then, the $\text{Bi}_2\text{Se}_3$-PVA and $\text{Bi}_2\text{Te}_3$-PVA solutions were placed in an ultrasonic bath for 10 min to make sure the $\text{Bi}_2\text{Se}_3$ and $\text{Bi}_2\text{Te}_3$ powder fully binds with the PVA. After that, both TI suspensions are carefully poured onto petri dishes to avoid trapping any air bubbles and are left to dry at room temperature for 48 h to form $\text{Bi}_2\text{Se}_3$-PVA and $\text{Bi}_2\text{Te}_3$-PVA composite films. The thickness of $\text{Bi}_2\text{Se}_3$ and $\text{Bi}_2\text{Te}_3$ films is about 55 and 68 $\mu$m, respectively.

Figures 1(a) and 1(b) show the field emission scanning electron microscopy (FESEM) images of $\text{Bi}_2\text{Se}_3$ and $\text{Bi}_2\text{Te}_3$ film, respectively. As shown in Fig. 1(a), the $\text{Bi}_2\text{Se}_3$ film has a high density of microrods and micrograins, which can be clearly viewed and distributed randomly on the substrate.
surface. These microrods and micrograins are in irregular shapes with an average size of 0.3 to 1.66 μm. As shown in Fig. 1(b), the Bi₂Te₃ film has also a high density of acicular grains formation, such as micronetworks, which are in irregular shapes in a size between 0.5 and 1.9 μm.

Figure 2 illustrates the absorbance spectrum of the prepared PVA only, Bi₂Se₃-PVA and Bi₂Te₃-PVA films in the range of 200 to 1100 nm. The figure attests a constant absorbance for both Bi₂Se₃-PVA and Bi₂Te₃-PVA film, which indicates that they possess a broadband resonance wavelength, such as graphene. It is shown that the incorporation of Bi₂Se₃ and Bi₂Te₃ nanoparticles into the PVA increases the loss of the film. The Bi₂Te₃ film is slightly thicker and thus it has a higher loss compared to the Bi₂Se₃ film. However, at around 1100 nm, both Bi₂Se₃ and Bi₂Te₃ films suffer an almost identical absorption loss. It also shows that PVA polymer is an excellent host material for fabricating high-performance TI SA and not affected by this host polymer. Figure 3 shows the Raman spectra of the fabricated films. For Bi₂Se₃-PVA film, the three distinct peaks observed at ~67, ~126, and ~170 cm⁻¹ can be assigned to the A₁₇, E₂¹, and A₂₂ vibrational modes, respectively. It also shows the Raman spectrum of the developed Bi₂Te₃-PVA film with low peak at ~67 cm⁻¹ (A₁₇), ~96 (E₂¹), and 118 (A₂₂), respectively. The low peak is maybe due to the low concentration of the Bi₂Te₃ powder in PVA and the effect of the film thickness. We use the same ratio of Bi₂Se₃ and Bi₂Te₃ of 14 mg in 3 ml PVA, but Bi₂Te₃ powder has higher molecular weight (800.76 g/mol) compared to Bi₂Se₃ (654.84 g/mol). To improve the Raman spectrum of B₁₂Te₃-PVA film, we could increase the weight of Bi₂Te₃ powder. The simple approach in this work to mix the Bi₂Se₃ and Bi₂Te₃ powder with PVA at room temperature for scalable production SA is very attractive.

Fig. 1 FESEM images of (a) the Bi₂Se₃ and (b) Bi₂Te₃ composite film. Inset of each image represents a high magnification of the respective image.

Fig. 2 Linear transmission of the free-standing Bi₂Se₃-PVA and Bi₂Te₃-PVA films.

Fig. 3 Comparison of Raman spectrum of the free-standing Bi₂Se₃-PVA with Bi₂Te₃-PVA film.

Fig. 4 (a) Schematic of the nonlinear absorption measurement, (b) characteristics of the Bi₂Se₃-PVA film, and (c) characteristic of the Bi₂Te₃-PVA film.
The nonlinear optical response property for the Bi$_2$Se$_3$ and Bi$_2$Te$_3$ was investigated to confirm their saturable absorption by applying twin-detector techniques. Figure 4(a) shows the schematic of the nonlinear absorption measurement. The pulse input source used a mode-locked fiber laser with 1550-nm wavelength, which has femtosecond output pulse with a 17-MHz repetition rate, and a 900-fs pulse duration, in which the output power is ~5 mW. The laser was connected to the low-dispersion erbium-doped fiber amplifier, a variable optical attenuator, and then to a 3-dB optical coupler. The output power of the twin-detector was measured using two optical power meters (OPMs). The first OPM was connected to one port of a coupler and the second port of the coupler was connected to the Bi$_2$Se$_3$/Bi$_2$Te$_3$ film sample and then connected into the second OPM.

Figures 4(b) and 4(c) show the recorded data of transmissions at various input intensities for the Bi$_2$Se$_3$ and Bi$_2$Te$_3$ SAs, respectively, which were fitted by using the following simple saturable absorption equation:

\[ T(I) = 1 - a_0 \exp(-I/I_{sat}) - a_{ns}, \]

where \( T(I) \), \( a_0 \), \( I \), \( I_{sat} \), and \( a_{ns} \) stand for the transmission, modulation depth, input intensity, saturation intensity, and nonsaturable absorption, respectively. For Bi$_2$Se$_3$ SA, the modulation depth, nonsaturable absorption, and saturation intensity were measured to be ~15.9%, 77%, and 50 MW/cm$^2$, respectively, as shown in Fig. 4(b). As well as in Bi$_2$Te$_3$ SA, shown in Fig. 4(c), the modulation depth, nonsaturable absorption, and saturation intensity were obtained to be ~30%, 55%, and 40 MW/cm$^2$, respectively. In this case, the modulation depth of Bi$_2$Te$_3$ sample is higher; however, the saturation intensity is smaller than in the Bi$_2$Se$_3$ sample.

3 Experimental Setup

The schematic experimental setup of our Q-switched YDFL with Ti-based SA is shown in Fig. 5. The ring laser consists of a 1.5-m-long YDF as the active medium, a 980/1064-nm wavelength-division multiplexer (WDM), an optical isolator, a Ti:Bi$_2$Se$_3$ or Ti:Bi$_2$Te$_3$ SA, and a 10-dB output coupler. The YDF used has ytterbium ions concentration of 1500 ppm, a core diameter of 4 $\mu$m, numerical aperture of 0.20, and cut-off wavelength of 950 nm. It is pumped by a 980-nm laser diode through the WDM. The other end of the YDF is spliced to the 10-dB output coupler, which allows 10% of the output to be extracted for analysis. The remainder is channeled to the SA device and kept in the cavity to oscillate for laser generation. The SA device is fabricated by sandwiching a ~1-mm x 1-mm piece of the Bi$_2$Se$_3$ or Bi$_2$Te$_3$ composite film between two fiber ferrules using a fiber adapter. Index matching gel is applied at the connection to minimize parasitic reflections. The SA is connected to the isolator, which prevents backward reflection and ensures unidirectional operation. The other port of the isolator is spliced to the 1064-nm port of the WDM to link the ring cavity. The output laser is analyzed by an optical spectrum analyzer with a spectral resolution of 0.02 nm, an OPM, and a 2-GHz photodetector together with a-350 MHz oscilloscope or 7.8-GHz radio-frequency (RF) spectrum analyzer. The total cavity length is around 11 m.

4 Results and Discussion

The performance of the YDFL cavity was investigated by varying the power of the 980-nm pump. The continuous wave (CW) laser started as the power hit the threshold of 43.7 mW. Further, a self-starting and stable Q-switched operation was obtained when the pump power reached the threshold of 67.6 and 72.8 mW for Bi$_2$Se$_3$ and Bi$_2$Te$_3$, respectively. Figure 6 shows the output optical spectra of the Q-switched YDFL constructed with Bi$_2$Se$_3$ and Bi$_2$Te$_3$-based SA at 88.3-mW pump power. As shown, the output optical spectra of Bi$_2$Se$_3$ and Bi$_2$Te$_3$-based Q-switched YDFLs center at wavelengths of 1057.8 and 1043 nm with 3-dB bandwidth of 4.4 and 2 nm, and peak output power of ~30, and ~28 dBm, respectively. The CW YDFL output spectrum, which was obtained by removing the SA, is also shown for comparison. Without the SA, the YDFL operates at the center wavelength of 1062 nm. The operating wavelength shifts to a shorter wavelength with the employment of SA due to the film insertion loss. The Q-switched lasers operate at a shorter wavelength, which has a higher gain to compensate for the loss. This result also indicates that the Bi$_2$Te$_3$ film has a slightly higher loss than Bi$_2$Se$_3$. Spectral broadening is also observed in the Q-switched lasers due to the self-phase modulation effect in the ring cavity. The same ratio of Bi$_2$Se$_3$ and Bi$_2$Te$_3$ of 14 mg in 5 ml PVA was used, however, Bi$_2$Te$_3$ powder has a slightly higher molecular weight (800.76 g/mol) compared to Bi$_2$Se$_3$ (654.84 g/mol). The
Bi$_2$Se$_3$ powder has a higher degree of dispersion in PVA compared to Bi$_2$Te$_3$ powder and this will affect the performance of the developed film as a saturable absorber. Therefore, the bandwidth of the output laser of Bi$_2$Se$_3$ is much wider than Bi$_2$Te$_3$ as shown in Fig. 6.

The comparison performance of the Q-switched laser with Bi$_2$Se$_3$ and Bi$_2$Te$_3$-based SA is summarized in Fig. 7. A stable Q-switching pulses train is generated by increasing the pump power above 67.6 until 88.3 mW for Bi$_2$Se$_3$ SA, while from 72.8 until 98.4 mW for Bi$_2$Te$_3$ SA. Figure 7(a) shows the typical oscilloscope trace at the threshold pump power of both SAs. For Bi$_2$Se$_3$ SA, the typical oscilloscope shows at 67.6 mW and threshold pump power, which shows a pulse repetition rate of 48.83 kHz. The pulse train has an average output power of 0.65 mW, which matches the pulse energy of 13.3 nJ. With Bi$_2$Te$_3$ SA, the typical oscilloscope trace is shown at the threshold pump power of 72.8 mW, which has a repetition rate of 43.25 kHz and pulse width of 5.65 µs. Figures 7(b)–7(c) compare the pulse train and the envelope of the single pulse at maximum pump power of Q-switching pulses obtained by the Bi$_2$Se$_3$ and Bi$_2$Te$_3$ SA at a pump power of 88.3 and 98.4 mW, respectively. The repetition rate between two pulses is measured to be around 102 and 93.63 kHz, whereas the maximum Q-switching pulse envelope with a period of 3.48 and 2.01 µs as measured from the full-width half maximum. There is no amplitude fluctuation for the pulse within the tunable range of the repetition rate for both SAs.

Figure 7(d) shows the measured pulse repetition rate and pulse width against the pump power for both Q-switched lasers. It is shown that the pulse repetition rate linearly increases, while the pulse width linearly decreases with the pump power. This matches well with the passive Q-switched theory based on SA. The pulse repetition rate of Bi$_2$Se$_3$ SA-based Q-switched YDFL linearly increases from 48.83 to 102 kHz, while the pulse width decreases from 8.9 to 3.48 µs as the pump power rises from 67.6 to 88.3 mW. Similarly, the pulse width of the Bi$_2$Te$_3$-based Q-switched YDFL decreases from 5.65 to 2.01 µs with the increase in pump power from 72.8 to 98.4 mW. The associated repetition rate increases from 43.25 to 93.63 kHz within the same range. This is typical of a Q-switched fiber laser due to the gain compression effect in the laser cavity. Compared to the Bi$_2$Se$_3$ laser, the Bi$_2$Te$_3$ Q-switched laser shows a narrower tuning range for both repetition rate and pulse width. The pulse width YDFL of the Bi$_2$Se$_3$ is expected to drop further if the pump power can be increased more, as long as the damage threshold of the SA is not exceeded. The Q-switched threshold is also lower for the Bi$_2$Te$_3$ laser due to the thicker film layer, which allows more absorption. It is observed that the Q-switching pulse disappears when the pump power is increased above 88.3 and 98.4 mW for Bi$_2$Se$_3$ and Bi$_2$Te$_3$ SA, respectively. This is due to the SA, which stayed in the saturated state because of the continuous exposure to the high intensity of light, and there is no time for de-excitation. However, the two Q-switching lasers are obtained again by reducing the pump power within the range of operation.

Figure 7(e) shows the measured average output power and the calculated pulse energy of both Q-switched YDFLs as a function of pump power. As seen in the figure, the average output power increases with the pump power, while the pulse width decreases from 5.65 to 2.01 µs as the pump power is increased from 67.6 to 98.4 mW for Bi$_2$Te$_3$ and Bi$_2$Se$_3$ SA, respectively. The calculated pulse energy of Bi$_2$Se$_3$ is 13.3 nJ. With Bi$_2$Te$_3$ SA, the pulse width decreases from 5.65 to 2.01 µs as the pump power is increased from 67.6 to 98.4 mW, respectively. This is due to the SA, which stayed in the saturated state because of the continuous exposure to the high intensity of light, and there is no time for de-excitation. However, the two Q-switching lasers are obtained again by reducing the pump power within the range of operation.

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Fig. 7 The performance of Q-switched YDFL with Bi$_2$Se$_3$ and Bi$_2$Te$_3$-based SA: (a) pulse train at threshold pump power of 67.6 and 72.8 mW, respectively, (b) and (c) pulse train and single envelope pulse at maximum pump power of 88.3 and 98.4 mW, respectively, and (d) pulse width and repetition rate characteristics at various pump power. (e) The average output power and pulse energy characteristics at various pump power.
energy fluctuates within the pump power for both Q-switching lasers. For Bi$_2$Se$_3$-based YDFL, the output power increases from 0.65 to 1.27 mW within the pump power range from 67.6 to 88.3 mW. The maximum output power of 1.27 mW is obtained at the maximum pump power of 88.3 mW, while the maximum pulse energy of 16.6 nJ is recorded at 72.8 mW. In comparison, the average output power of the Bi$_2$Te$_3$-based YDFL is obtained at 72.8 mW. In comparison, the average output power of the Bi$_2$Te$_3$-based YDFL is obtained at 83.2 mW pump power.

Figure 8 shows the RF spectra of the Q-switched YDFLs configured with Bi$_2$Se$_3$ and Bi$_2$Te$_3$, which were achieved with a resolution bandwidth of 3 kHz at the maximum pump powers of 88.3 and 98.4 mW, respectively. Both spectra were taken using an RF spectrum analyzer via a 1.2-GHz photodetector. The maximum repetition rate of 102 kHz and 93.63 kHz for the Bi$_2$Se$_3$ and Bi$_2$Te$_3$ SA, respectively. Both SAs show good Q-switching performance in the 1-μm wavelength region.

5 Conclusion

This paper presents details of a passively Q-switched YDFL based on Bi$_2$Se$_3$ and Bi$_2$Te$_3$ nanoparticles SA operating in the 1-μm region. Self-starting and stable Q-switched pulse trains with peak wavelength centered at 1057.4 and 1044.5 nm were obtained. The pulses have a shortest pulse duration of 3.48 and 2.01 μs, maximum pulse energy of 16.6 and 29.95 nJ, and pulse frequencies from 48.83 to 102 kHz and 43.25 to 93.63 kHz for the Bi$_2$Se$_3$ and Bi$_2$Te$_3$ SA, respectively. Both SAs show good Q-switching performance in the 1-μm wavelength region.

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Passively mode-locked ytterbium-doped fiber laser operation with few layer MoS$_2$ PVA saturable absorber

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Abstract

A passive mode-locked Ytterbium-doped fiber laser (YDFL) is experimentally demonstrated using a multi-layered molybdenum disulfide (MoS$_2$) as a Saturable Absorber (SA). The MoS$_2$ film was obtained based on a liquid phase exfoliation of MoS$_2$ crystal. A small slice of MoS$_2$ film was sandwiched in between two optical ferrules via a fiber adapter and linked into an YDFL cavity to generate a stable mode-locked operation at a wavelength of 1073.86 nm with repetition rate of 18.8 MHz. At pump power of 202.9 mW, the maximum pulse energy of 0.1 nJ was achieved with an output power of 1.84 mW. The MoS$_2$ film was fabricated through a simple process and it has a modulation depth of 9.7% and saturating intensity of 40 MW/cm$^2$.

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1. Introduction

In the recent years, mode-locked fiber lasers have extensively engaged interest due to their application benefits, such as micromachining, communication, and optical systems; as well as their nature being compact, cost-effective and easy to setup [1,2]. Active mode-locking pulse train can be achieved by inserting electronic switching such as acousto-optic modulator [3]. Passively mode-locked fiber lasers are also widely explored in recent years by utilizing saturable absorption of optical materials. Various saturable absorbers (SAs) have been demonstrated so far such as semiconductor saturable absorption mirror (SESAMs) [4,5], carbon nanotubes (CNTs) [6,7], and graphene [8,9].

Semiconductor saturable absorption mirrors (SESAM) are good SA device which can be used to generate a mode-locked fiber laser in both 1 μm and 1.5 μm regions [5,10]. However, its application is still limited due to their difficult fabrication methods, complex design, expensive cost and narrower wavelength operation range. Graphene and carbon materials have the advantages of low cost, easy fabrication and operable in a wide wavelength range. Mode-locking pulses have been demonstrated using graphene and carbon nanotubes (CNTs) as SA in the various fiber laser cavities based on erbium-[6,11,12], ytterbium- [13–15] and thulium- [16,17] doped fiber gain medium. The operation wavelength of CNT is related with diameter of the nanotubes. Graphene has a 2D zero-bandgap semiconductor material and low absorption co-efficiency, which gives it the advantages of wideband saturable absorption as well as ultra-fast recovery time. These advantages lead researchers to 2D materials. Topological insulators have also joined the 2D materials for ultra-fast fiber laser applications such as Bi$_2$Se$_3$, Bi$_2$Te$_3$ and many others [18,19].
To date, another 2D nanomaterial, molybdenum disulfide (MoS₂), is also drawing more interest especially for mode-locked fiber laser applications. The reason for this growing interest is due to its material’s thickness, which gives an excellent optical laser properties as well as the band-gap [20,21]. Until today, only a few works have been reported on MoS₂ based SA [20–24]. For instance, Zhang et al. demonstrated tunable mode-locked fiber laser that operates from 1535 to 1565 nm using MoS₂, which was obtained through liquid phase exfoliation [22]. Du et al. reported the generation of 656 ps pulse duration and repetition rate of 6.74 MHz which operated at 1042.6 nm wavelength based on a few layers MoS₂ SA, which fabricated through the conventional hydrothermal intercalation process [23]. Sathiyan et al. proposed mode-locked Ytterbium-doped fiber laser (YDFL) using few-layer MoS₂ prepared by mixing MoS₂ solution with polyvinyl alcohol (PVA) [24].

In this paper, we demonstrate a passively mode-locked YDFL using a few layered molybdenum disulfide (MoS₂) as a Saturable Absorber. The few layer of MoS₂ polymer is a composite of liquid phase exfoliation (LPE) of chemically pristine crystal. A small piece of MoS₂-PVA was inserted in the YDFL cavity, sandwiching it in between two fiber optical ferrules with a connector to obtain mode-locking pulses with repetition rate of 18.8 MHz and pulse energy of 0.1 nJ. In this work, the liquid phase exfoliation method was used to get the MoS₂ layer. This method is simple, cheap and does not require any complicated procedures.

2. Fabrication and characterization of the MoS₂ film

The liquid phase exfoliation (LPE) process for MoS₂ exfoliation was carried out in 2 steps. The bulk MoS₂ crystals were first mixed together with a solvent that gets a comparable surface energy with MoS₂ (∼75 mJ m⁻²) [25]. Likewise, Dimethylformamide (DMF) solvent was chosen with a concentration of 5 mg/ml for this work. Following, the second step was to exfoliate the material through a sonication process. In this process, local pressure variations were generated to sufficiently separate the atomic layers of the bulk crystal [26,27] in the solvent to produce a dispersion, which was rich in few-layer flakes [26]. Then, the MoS₂ dispersion was kept to be ultra-sonicated for 24 hours before it was centrifuged for 1 hour at ~3000 rpm. This allowed the un-exfoliated sediment to settle at the bottom since the larger flakes descended more rapidly than the exfoliated few-layer flakes in the centrifuge cell [28]. The top portion of the dispersion containing few-layer flakes was taken out for further composition fabrication process.

A few steps were followed in order to produce MoS₂ polymer film for this experiment. Firstly, ∼10 ml of decanted MoS₂ dispersion was put together with a 10 mg/ml concentrated ∼20 ml of polyvinyl alcohol (PVA) solution. This solution was a result of ∼200 mg of PVA being dissolved in 20 ml of deionized water. By this, 30 ml of MoS₂-PVA was prepared, which was then heated at a temperature of ∼80 °C and stirred with a magnetic object until the solution was brought down to an approximate volume of ∼10 ml. These series of actions took around 7 hours to be done, following which the resulting MoS₂-PVA solution was placed in a glass substrate and put in an oven for 4 hours at a temperature of ∼80 °C in order to dry and obtain a composite film with a thickness of ∼40 μm.

Field emission scanning electron microscopy (FESEM) and Raman spectroscopy were performed on the fabricated MoS₂-PVA film sample. Fig. 1a shows the FESEM image that illustrates a MoS₂ flakes, which were embedded into the PVA polymer film. The scanning electron microscopy (SEM) image for a single flake is shown in Fig. 1b, which was obtained from the prepared MoS₂ dispersion solution. It shows a lateral size of about 400 nm with thickness of around 5–8 layers. Fig. 1c presents the Raman spectrum recorded using a spectrometer at a 514 nm beam of an Argon-ion laser that was radiated on the film for 10 seconds with an exposure power of 10 mW. As shown in the figure, the sample displays two signature peaks at 383 cm⁻¹ and 407 cm⁻¹ which are related to in-plane vibration of Molybdenum and Sulfide atoms and out of plane vibration of Sulfide atoms respectively. The frequency difference was recorded to be 24 cm⁻¹. It was observed that the E₂g mode according to the in-plane motion shifted to a shorter wavelength after complete exfoliation. This indicated that the number of layers of MoS₂ were in the range of 2–5 layers [29]. Fig. 1d shows the Raman spectrum of a pure PVA solution, which shows a peak at 3000 cm⁻¹ region for comparison purpose.

A nonlinear absorption measurement was also performed on the fabricated MoS₂-PVA film to confirm its saturable absorption as shown in Fig. 2a. In the experiment, a stable mode-locked fiber laser operating at 1565 nm wavelength with repetition rate of 15 MHz and pulse width of 0.9 ps, was employed as the input pulse source. The mode locking signal results were then amplified using an erbium-doped fiber amplifier (EDFA) in order to obtain a high peak power output to efficiently saturate the MoS₂ sample. The output port of the amplifier was connected with a variable optical attenuator to control the launching output power to the system before it is launched into a 3 dB optical coupler and two optical power meters. The first optical power meter was linked to one port of coupler and the second port of coupler was linked to the MoS₂ film sample then connected into the second optical power meter. The resultant characteristic of MoS₂ film as SA is shown in Fig. 2b. The absorption power was recorded as a function of incident intensity on the device by modifying the input optical laser power. The experimental data for absorption were prepared depending on the two-levels of SA model [30].

\[
a(l) = \frac{\alpha_s}{1 + l/l_{sat}} + \alpha_{ns}
\]

where \(a(l), \alpha_s, l, l_{sat}\) and \(\alpha_{ns}\) are stand for absorption, modulation depth, input intensity, saturation intensity, and non-saturation loss respectively. For the MoS₂ sample that was used in this work, the modulation depth, non-saturable intensity, and saturation intensity were measured to be approximately 9.7%, 10.9%, and 40 MW/cm², respectively.
Fig. 1. (a) FESEM image of the MoS$_2$ film, (b) SEM image of the MoS$_2$ flake, (c) Raman spectrum of the MoS$_2$-polymer composite film (d) Raman spectrum of the pure PVA.

3. Experimental setup for the mode-locked YDFL

The proposed configuration of mode-locked YDFL with MoS$_2$ SA is shown in Fig. 3. It uses a 0.7 m long of Ytterbium-doped fiber (YDF) with 1300 dB/m peak absorption at 977 nm as a gain medium. The YDF was pumped by a 974 nm laser diode through a 980/1060 nm wavelength division multiplexer (WDM). An isolator is used in the ring cavity to avoid backward reflection as well as ensure unidirectional operation. The MoS$_2$ SA device was composed by sandwiching the fabricated MoS$_2$ PVA film in between two fiber ferrules via an adapter as shown in the inset of Fig. 3. It is incorporated into the laser cavity to act as a mode-locker for generating ultrashort pulses train. A 10 dB coupler is used to tap out the output laser while allowing 90% of the photons to oscillate inside the cavity. The cavity length is measured to be about 10.6 m with normal dispersion...
of 0.242 ps²/km nm at 1030 nm. The output spectrum of the laser is measured by an optical spectrum analyser (OSA) with a 0.02 nm spectral resolution while the temporal characteristic of the laser is investigated using an oscilloscope coupled with a 2-GHz photodetector. The radio-frequency (RF) spectrum is measured by an RF spectrum analyzer with a range operation from 9.0 kHz to 7.8 GHz.

4. Results and discussion

At first, we observed a continuous wave (CW) laser operation when the pump power reached to 58.0 mW. As the pump power was further increased to 78.7 mW, the self-started and stable mode-locked pulse train was generated. The mode-locked pulse train was maintained with further increase in pump power up to the maximum pump power of 202.9 mW. Fig. 4a shows the output spectrum of the mode-locked laser at pump power of 202.9 mW. The peak wavelength was centered at 1073.86 mW with 3 dB bandwidth of 0.06 nm without Kelly sideband, which indicates that the pulses operate in normal dispersion. The typical mode-locking pulse train at pump power of 202.9 mW is shown in Fig. 4b. It is observed that the peak to peak spacing of the pulse train was about 53 ns, which equals to the repetition rate of 18.8 MHz. The repetition rate obtained is corresponded with the fundamental frequency for cavity length of 10.6 m. As shown in the figure, the pulse width is measured to be 3.12 ns, which is a lot broader than the actual pulse width. This is attributed to the oscilloscope resolution limitation. To estimate the actual pulse width, we used mathematical calculation based on time bandwidth product (TBP). Assuming that the TBP is 0.441 for Gaussian pulse profile [31], minimum possible pulse width is calculated to be approximately 28.3 ps.

Fig. 4c shows the output power and pulse energy of the mode-locked laser at various pump powers. As seen in the figure, both output power and pulse energy linearly increased with the pump power. At the maximum pump power of 202.9 mW, the maximum output power and pulse energy are obtained at 1.84 mW and 0.1 nJ, respectively. Fig. 4d shows the RF spectrum for YDFL laser. The fundamental frequency of the laser is obtained at 18.8 MHz with a signal-to-noise ratio (SNR) of more than 30 dB, which indicates the stability of the mode-locking operation. It is worthy to note that there is no appearance of fundamental frequency noticed as the MoS₂ is taken off at all level of pump power.

5. Conclusions

A passive mode-locked YDFL was successfully demonstrated using a few layered MoS₂ PVA film as a mode-locker. The MoS₂ film was obtained based on a LPE of MoS₂ crystal. Stable and self-starting passive mode-locking pulses were achieved within the pump power of 78.7 mW to 202.9 mW. It operates at 1073.86 nm with a fixed repetition rate of 18.8 MHz with picoseconds pulse width. The maximum pulse energy was calculated to be approximately 0.1 nJ at pump power of 202.9 mW.
Fig. 4. Mode-locked YDFL performances. (a) Output spectrum and (b) Oscilloscope train at maximum pump power. (c) Output power and pulse energy. (d) RF spectrum.

This piece of work tackled an important aspect that contributes to study nonlinear optical properties of MoS2, which may in turn guide to future paths for ultrafast photonics applications.

References


Original research article

Mode-locked ytterbium-doped fiber laser using mechanically exfoliated black phosphorus as saturable absorber

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\textbf{A R T I C L E  I N F O}

Article history:
Received 5 July 2017
Received in revised form 3 August 2017
Accepted 3 August 2017

Keywords:
Mode-locking
Black phosphorus
Passive saturable absorber

\textbf{A B S T R A C T}

We experimentally demonstrated a passive, stable mode-locked ytterbium-doped fiber laser (YDFL) based on black phosphorus (BP) as saturable absorber (SA). The BP-SA was prepared by mechanically exfoliation of the crystal and transferred the acquired thin layer onto the end surface of a standard FC/PC fiber connector. By incorporating the BP-SA into the YDFL cavity, a stable mode-locked operation was obtained at 1033.76 nm wavelength with a repetition rate of 10 MHz. The maximum pulse energy of 2.7 nJ and peak power of 0.83 kW were obtained at the maximum pump power of 200 mW. The experimental results indicate that the BP SA can be used successfully to generate stable mode-locking pulse at 1 \( \mu \)m region.

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1. Introduction

Recently, ultra-short optical pulse sources have attracted widespread interests because of their useful applications in many areas such as micromachining, communication, and optical systems [1–3]. Passive mode-locking techniques are preferable due to their advantages such as nature being compact, low cost and easy to setup [4,5]. Furthermore, active mode-locking requires an additional switching electronic device such as acousto-optic modulators to be incorporated in the laser cavity [6]. Passively mode-locked fiber lasers were normally demonstrated by utilizing saturable absorption of optical materials. Various saturable absorbers (SAs) have been demonstrated so far such as semiconductor saturable absorption mirror (SESAMs) [7,8], carbon nanotubes (CNTs) [9,10], and 2D materials including graphene [11,12], topological insulators (TIs) and transition metal dichalcogenides (TMDCs) [13,14]. However, SESAM based SAs have a limited wavelength operation and complicated fabrication as well as costly. CNT commonly used as SAs because they are cheaper and simple in fabrication process, yet the diameters of the nanotubes define the range of operation wavelength. Graphene has a comparatively weak absorption of 2.3% per layer, which limit its applications while TIs based SA is difficult to be fabricated since it is a compound of two different elements. TMDCs have a large bandgap that is more suitable for visible band [15,16].

To date, another 2D nanomaterial, black phosphorus (BP) has also drawn the interest as a promising material for ultrashort pulse applications [17–20]. The reasons for this growing interest is because it has the most thermo-dynamically stable allotrope of phosphorus and a direct energy band structure in any case of its thickness, which can give excellent
optical laser properties [21,22]. Compared to other 2D materials, BP has a direct band gap changing from 0.3 to 2.0 eV in BP layer. The advantage of bandgap is helpful for its optoelectronics applications. This makes the material more suitable for nonlinear short pulse modulation and thus it can be used as a SA for generating mode-locking pulses [16,23]. In this paper, we demonstrate mode-locking pulses generation in Ytterbium-doped fiber laser (YDFL) cavity by using a newly developed BP based SA. The SA was fabricated by mechanically exfoliating a BP layer from a bulk crystal. The BP layer was transferred onto the end surface of a FC/PC fiber ferrule, with was then mated to another ferrule to construct an all-fiber SA device. By integrating the SA into an YDFL cavity, a mode-locked laser was realized to operate at 1033.76 nm wavelength with 10 MHz repetition rate and 2.7 nJ pulse energy.

2. Preparation and characterization of BP SA

In our experiment, the multi-layer BP SA was prepared by a mechanical exfoliation method as described in Fig. 1. The similar techniques have been generally employed for generating pulses for other 2D nanomaterials such as graphene and TIs [24,25]. This is due to its reliability and simple fabrication process, which do not have any chemical procedures. At first, we moderately peeled off thin flakes from a big block of commercially available BP crystal (purity of 99.995%) using clear scotch tape. Then, we repeatedly pressed the flakes adhered on the scotch tape to produce a thin layer of BP, which is sufficiently thin enough to transmit light with a high efficiency. A fresh standard FC/PC fiber end surface was pushed down onto the BP layer to transfer the material into the ferrule. With the aid of index matching gel, the ferrule was then connected with another FC/PC fiber ferrule using a standard flange adapter to construct an all-fiber BP based SA device.

Fig. 2(a) shows the Field Emission Scanning Electron Microscopy (FESEM) image, which confirms the existence of uniform multi-layer phosphorus and the absence of >1 μm aggregates or voids in the tape, which otherwise result in non-saturable scattering losses. The composition of the multi-layer BP tape is examined by the energy dispersive spectroscopy (EDS) on the FESEM as shown in Fig. 2(b). The presence of BP material on the scotch tape adhesive surface was confirmed by the

![Fig. 1. The fabrication process of mechanical exfoliated BP based SA.](image1)

![Fig. 2. (a) FESEM image, and (b) EDS data from the FESEM image of the prepared BP tape, which confirmed the present of phosphorus.](image2)
presence of the high peak of phosphorus in the spectroscopy. We also performed Raman spectroscopy on the prepared BP tape sample. Fig. 3 shows the Raman spectrum, which is recorded by radiating an Argon ion laser onto the tape for 10 ms with an exposure power of 10 mW. As shown in the figure, the sample exhibits three distinct Raman peaks at 360 cm\(^{-1}\), 438 cm\(^{-1}\) and 465 cm\(^{-1}\), corresponding to \(A_{1g}\), \(B_{2g}\) and \(A_{2g}\) vibration modes of a layered BP. While \(B_{2g}\) and \(A_{2g}\) modes correspond to the in-plane oscillation of phosphorus atoms in BP layer, the \(A_{1g}\) mode corresponds to the out-of-plane vibration.

The nonlinear optical response property for the BP SA tape was then experimentally investigated using an absorption technique in order to confirm its saturable absorption level. We used an input light source from a homemade of mode-locked fiber laser operating at 1557 nm wavelength with 1.5 ps pulse width and 17.4 MHz repetition rate. The transmitted power and also a reference power for normalization are recorded as a function of incident intensity on the tape by varying the input laser power. With increasing peak intensity, the material absorption decreases as shown in Fig. 4, confirming the saturable absorption characteristic of the multi-layer BP. The experimental data for absorption are fitted according to a simple two-level SA model [26]:

\[
\alpha(I) = \frac{\alpha_s}{1 + I/I_{sat}} + \alpha_{ns}
\]

where \(\alpha(I)\) is the absorption, \(\alpha_s\) is the modulation depth, \(I\) is the input intensity, \(I_{sat}\) is the saturation intensity, and \(\alpha_{ns}\) is the non-saturable absorption. As shown in Fig. 4, the modulation depth, non-saturable intensity, and saturation intensity are 7.5%, 57.5% and 0.35 MW/cm\(^2\), respectively. Taking into account its nonlinear optical response leading to absorption saturation at relatively low fluence, the mechanically exfoliated BP meets basic criteria of a passive SA for fiber lasers.
is worthy to note that a laser beam of 100 mW was injected into the BP that is deposited onto the fibre ferrule. No optical damage is observed and this shows that the produced SA has high optical damage.

3. Laser configuration

In the experiment, we investigate the employment of BP SA for ultrafast laser generation in YDFL cavity to operate at 1 μm region. Fig. 5 shows the experimental setup, which mainly consists of Ytterbium-doped fiber (YDF) with length of about 1.5 m as a gain medium. The YDF (Yb1200-4/125) has a core and cladding diameters of 4 μm and 125 μm respectively, with a numerical aperture of 0.20, as well as the cut-off wavelength of 1010 nm and ion absorption of 280 dB/m at 920 nm. In order to ensure unidirectional operation, an isolator is spliced into the cavity. The 980 nm laser diode (LD) is coupled into the fiber ring resonator with a 980/1064 nm wavelength division multiplexer (WDM). A 10 dB output coupler was used in the resonator to extract 10% of the output for analysis while allowing 90% of the light to oscillate in the laser cavity. A polarization controller (PC) was incorporated inside the laser cavity between the SA device and isolator to adjust the polarization state of the oscillating light and optimizes the laser mode-locking performance. The output laser is analyzed by an optical spectrum analyzer (OSA) with a spectral resolution of 0.02 nm, an optical power meter and a 2 GHz photodetector together with a 350 MHz oscilloscope or 7.8 GHz radio-frequency (RF) spectrum analyzer. The laser cavity consists of 1.5 m ytterbium-doped fiber (YDF) with a group velocity dispersion (GVD) of 24.22 ps²/km and a single-mode fiber (SMF, HI 1060) with a total length of 19.5 m with GVD of 21.91 ps²/km as the other fiber component. The net cavity dispersion was normal with a value estimated to be 0.46 ps².

4. Results and discussion

At first, the laser cavity was tested without incorporation of the SA device to confirm that there is no pulse obtained on the oscilloscope. Then the prepared BP SA is incorporated into the laser cavity as a mode-locker. As the pump power reaches 123.7 mW threshold, a continuous-wave laser was firstly obtained. A stable mode-locking pulse was obtained as the pump power is further increased to 130.8 mW. The pulses train operating at repetition rate of 10 MHz was easily achieved by carefully controlling the rotation of the PC as the pump power is fixed within 130.8 mW–200 mW. Fig. 6 shows output spectrum of the mode-locked pulses at pump power of 200 mW. The spectrum shows a central wavelength of 1033.76 nm and 3 dB spectral bandwidth of 0.48 nm with no Kelly sideband, which confirmed the operation of laser in normal dispersion.

Fig. 6. Optical spectrum of the proposed mode-locked YDFL at pump power of 200 mW.

Fig. 7. Typical pulse train of mode-locking YDFL at two different pump powers.
The narrow spectral bandwidth is due to the minimal nonlinearity and high net cavity dispersion [27]. The Q-switching operation was also realized in the YDFL with the BP SA by increasing the cavity loss. It was achieved by replacing the 90/10 output coupler with a 3 dB coupler. The Q-switched laser has a pump threshold of 55 mW, a pulse repetition rate that is tunable from 8 to 33 kHz, and the narrowest pulse width of around 10 μs.

Fig. 7 shows the typical pulses train trace of the mode-locked YDFL measured by oscilloscope with pulse period of 96 ns at two different pump powers of 151.5 mW and 200 mW. It indicates there is no fluctuations in the pulse train. The achieved repetition rate pulse of 10 MHz is matched with the cavity length of 21 m. As shown in the same figure with the limitation of the oscilloscope resolution, the achieved pulse width of 49 ns is much broader than the actual pulse width. As an alternative, the pulse width can be measured by using calculation based on the time bandwidth product (TBP) formula. A mathematically estimated a minimum possible pulse width of approximately 3.27 ps with TBP of 0.441 for Gaussian pulse profile and 3 dB spectral bandwidth of 0.48 nm. The radio frequency (RF) spectrum for our YDF laser is shown in Fig. 8, which has confirmed that the mode-locked has a fundamental frequency of 10 MHz with a signal-to-noise ratio (SNR) up to 39 dB. By removing out the BP SA from the ring cavity, the mode-locking operation could not be achieved, no matter how we adjust the PC and

![Graph showing variation in output power, pulse energy, and peak power at different pump power.](image-url)
pump power. This verified that the mode-locking operation is purely caused by the BP SA. These results show that black phosphorus and other similar 2D materials may have a good potential for application in ultrafast photonics.

Fig. 9 shows the graphs of an average output, pulse energy and peak power as a function of input pump power for the mode-locked YDFL. As seen, the average output power, pulse energy, peak power rise linearly with the pump power. The average output power is increased from 4.7 to 27 mW as the pump power is increased from 130.8 to 200 mW as shown in Fig. 9(a). The pulse energy is measured based on the output power and repetition rate as shown in Fig. 9(b). The maximum pulse energy is 2.7 nJ at the maximum pump power of 200 mW. Fig. 9(c) shows the peak power against the pump power, which the maximum peak power is 0.83 kW at 200 mW pump power. In order to examine the long term stability of the mode-locked laser, we continuously monitor the output signal by an optical spectrum analyzer and an oscilloscope for 48 h. It is observed that central wavelength and 3 dB bandwidth of the spectrum are almost unchanged while the repetition rate stayed constant throughout the experiment. This observation proves the stability of BP as a SA in mode-locked fiber laser.

BP has a direct transition for all thicknesses, which shows a significant benefit for optoelectronic applications, especially at the long-wavelength range. As it can offer a broad tuning range for a bandgap with number of layers, and can bridge nearly the entire range between the zero bandgap of graphene and the large bandgap of TMDCs. Furthermore, the preparation of the BP is also simple and thus it is preferable compared to TIs, which limit themselves by the complex fabrication processes. However, the BP materials are very hydrophilic and thus the SA is easily damaged with the exposure to oxygen and water molecules. Nevertheless, the BP SA remain undamaged as we tuned pump power up to 200 mW for more than 2 days. By keeping in an air tight container, less expose to oxygen or water molecules helps to protect BP SA from damage easily. The experimental results verify the mode locking ability of the newly developed BP based SA. This shows that the BP could be used to develop promising optoelectronic devices with high power tolerance, offering inroads for more practical applications, such as large energy laser mode-locking, nonlinear optical modulation and signal processing etc.

5. Conclusion

Passively mode-locked YDFL using a multi-layer BP based SA is demonstrated. The BP SA was prepared by mechanical exfoliation method before its layer is transferred onto the end surface of a standard FC/PC fiber connector to form a fiber-compatible SA. The proposed mode-locked YDFL operates at 1033.76 nm wavelength with a repetition rate of 10 MHz. The highest pulse energy and peak power is measured to be around 2.7 nJ and 0.83 kW respectively at pump power of 200 mW. The fabrication of BP SA is easy, cheap and suitable device to generate mode-locked laser in 1 µm wavelength region. Our experimental results suggest that a multi-layer BP is a promising material for mode-locked laser applications.

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