A 2.95 GHz, femtosecond passive harmonic mode-locked fiber laser based on evanescent field interaction with topological insulator film

Peiguang Yan,1 Rongyong Lin,1 Shuangchen Ruan,1* Aijiang Liu1 and Hao Chen1

1 Shenzhen key laboratory of laser engineering, Shenzhen University, ShenZhen, 518060, China
*scruan@szu.edu.cn

Abstract: By utilizing the pulsed laser deposition (PLD) method, we fabricated a kind of microfiber-based topological insulator (TI) saturable absorber (SA) which has inherent merits of effective and robust properties. We also proposed a newly explanation for the impact of nonlinear effect of SA on the harmonic mode-locking (HML) behavior. Upon employing on the SA, we achieved stable fundamental mode-locking (FML) at central wavelength of 1562.4 nm with pulse duration as short as 320 fs. By adjusting the intracavity polarization state at maximum pump power of 395 mW, we obtained stable femtosecond harmonic soliton pulse generation with repetition rate of 2.95 GHz and output power of 45.3 mW. Our results demonstrated that the microfiber-based TI PLD film SA is a promising device for practical multi-GHz ultrashort pulses generation.

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OCIS codes: (140.4050) Mode-locked lasers; (160.4330) Nonlinear optical materials; (140.3510) Lasers, fiber.

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1. Introduction

New and high-performance saturable absorbers (SAs) are always among the hottest research topics in ultrafast photonics. To date, the reported SAs are involved in semiconductor saturable absorber mirrors (SESAMs) [1], gold nanoparticles [2, 3], carbon nanotubes (CNTs), graphene series [4–27], topological insulator (TI), and few-layer transition-mental dichalcogenides [28, 29] such as MoS$_2$ and WS$_2$. Although SESAMs have been well-developed and applied in commercial lasers, they often have relatively narrow operation...
bandwidth and complicated manufacturing technology. The gold nanoparticle has large third-order nonlinearity but the recovery time is in few picoseconds. CNTs and graphene have been intensively studied in passive mode-locked fiber laser as SA materials due to easy fabrication and fast recovery time. Unlike CNTs, Graphene has a Dirac-like electronic band structure with zero-bandgap and exhibits broadband saturable absorption property. Recently, topological insulators (TIs) have emerged as a new series of SAs with characteristics of a small band gap in the bulk state and a gapless metallic state in the edge/surface [30–32]. Noteworthy, TIs also possess broadband wavelength operation characteristic and giant third order nonlinear property [33, 34]. Therefore, TIs have attracted much attention for generating short and high-energy laser pulses [35–38].

Up to now, various fabrication methods of TI SA have been reported including of the molecular beam epitaxy (MBE) method, the mechanical exfoliated method and the liquid-phase exfoliation method, the bulk-structured TI method [39–48]. Usually, the MBE method is limited by the expensive equipment, and the growth is usually on bulk substrate. The mechanical exfoliation method has a low repeatability. The liquid-phase exfoliation method can fabricate nanostructured TIs. But in the actual application, the nanostructured TIs should be hosted on the fiber ferrule or doped into thin polyvinyl acetate (PVA) film, then sandwiched between two fiber connectors in laser system. However, this type of SA device usually has short interaction length and low power tolerance. Another approach is to deposit the nanostructured TI on side surface of microfiber, however, this reported SA device still suffers from a large insertion loss (IL: ~5.23 dB) and high nonsaturable loss (~69.9%) [49]. For the bulk-structured TI method, the TI should be polished smooth, then attached on the side-polishing fiber (SPF). In addition, the index match oil is needed to induce the evanescent wave interaction with TI layer. Hence, the bulk-structured TI/SPF device is not suitable for package in the compact laser system.

As we know, pulsed laser deposition (PLD) is a mature thin film deposition technique, and suitable for mass production. By changing the deposition parameters, including laser pulse energy, temperature and depositing time [64], the film thickness can be controlled. On the other hand, microfiber has a simple fabrication process based on the mature fused-taper method compared with SPF. If coating the TI film on side surface of microfiber via PLD method, it can be predicted that the SA device would have the following inherent merits compared with the common sandwiched method: 1) Effective, this type of SA device has longer interaction length and higher power tolerance, which are the inherent properties of SA devices based on the evanescent wave interaction. 2) More Robust, the PLD film is not easily disaffiliated because the film is totally growth on the side surface of microfiber. If further packaged by heatshrink tube, the film can be fully protected. 3) Suitable for mass production, the fabrication process is simple and suitable for mass production based on the mature fused-taper method and PLD method. However, according to our knowledge, no research has been carried out for fabricating SA (including the graphene and TI) on microfiber by PLD method.

High repetition rate pulse fiber lasers have applications in optical communication, astronomical frequency combs, and laser spectroscopy. Passively harmonic mode-locking (HML) is regarded as a very simple way to multiply the repetition rate up to GHz regime without reducing the laser cavity length. According to the soliton area theorem [50], the peak power-limiting effect causes the multiple pulsing [51–56] of quantized soliton per round trip as the pump power increases. So far, the reported highest repetition rate of passive HML fiber laser was 22.2 GHz based on nonlinear polarization rotation (NPR), but the laser had a low pump conversion efficiency and a <30 dB supermode suppression ratio (SMSR) [57, 58]. In real SAs, till now the reported HML based on CNT and graphene were 4.915 GHz and 2.22 GHz, respectively [59–63]. However for HML based on TIs, the maximum repetition rate was 2.04 GHz with pulse duration in few-picosecond regime, and output power at a level of ~5 mW. For a brief comparison, the detailed values of their performances were also summarized in Table 1.
In this paper, a kind of microfiber-based TI SA was firstly reported based on the PLD method. It was found that this SA had interesting nonlinear saturable property owing to the strong nonlinear effect induced by microfiber and TI film. Upon employing on this SA, we achieved stable fundamental mode-locking (FML) at central wavelength of 1562.4 nm with pulse duration as short as 320 fs. By adjustment the intracavity polarization state at the maximum pump power, we also obtained stable HML with repetition rate up to 2.95 GHz and output powers beyond 45 mW. Our results revealed that the microfiber-based TI film SA is a promising device for practical multi-GHz ultrashort pulses generation.

2. Fabrication and characterization of microfiber-based Bi$_2$Te$_3$ film SA

The fabrication process could be divided into two steps: fused-tapering and PLD coating. In step 1, the SMF-28 fiber was tapered to microfiber. The tapered waist of microfiber, marked by the arrow in Fig. 1(a), had a length of 0.6 mm and a diameter of ~54 μm. In step 2, the high energy Nd:YAG laser (SL II-10, Surelite) was operated with 2 mJ/pulse (2 W average power, and 10 Hz repetition rate). The laser beam was focused on the Bi$_2$Te$_3$ target inside a vacuum chamber with vacuum degree of $5 \times 10^{-4}$ pa. The inspired plasma plume was deposited on microfiber within 90 minutes. To avoid pollution from the plastic polymers of fiber, the deposition temperature was fixed at room temperature. Figure 1(b) was the leaky field observed in splicer (Fujicura, FSM-60S) by injection of supercontinuum light (Ylphotonics, SC-5-CFS), where the Y view was the face toward plasma plume while the X view was the sheltered face. It evidenced that the light could really penetrate into the TI film, which in turn gave a modulation on light along the TI-film-coated taper region. Additionally, it demonstrated that the filmy thickness was not a constant. Figure 1(c) showed this thin layer of TI film measured by scanning electron microscope (SEM), which stacked tightly on side surface of microfiber with an inherent robust feature. The thickest location was ~670 nm. Figure 1(d) was one part of the enlarged regions of TI film, as marked in Fig. 1(c). It can be seen that the filmy surface had an appearance like tiny crystal grains, which stacked compactly with irregular size. The X-ray diffraction (XRD) pattern of Bi$_2$Te$_3$ film, as shown in Fig. 1(e), exhibited high [006, 0015] orientations and some common characteristic peaks with the bulk Bi$_2$Te$_3$. Unlike the SPF/TI SA, our microfiber-based TI-film SA can be packaged by heatshrink tube to form a practical SA device, as shown in the inset. The insertion loss (IL) of this SA device was measured ~1.23 dB@1550nm.

The nonlinear saturable transmission measurement was carried out using an in-house made femtosecond laser source (central wavelength: 1560.8 nm, repetition rate: 18.55 MHz, pulse duration: ~140 fs). The measuring method was similar to that in our previous work [26]. The following formula was used to obtain the fitting curve:

\[
\text{Intensity} = I_0 \left(1 - \frac{I}{I_0} \right)^n
\]
\[ T(I) = 1 - \Delta T \times \exp \left( -\frac{\Delta T}{I_{sat}} \right) - T_{ns} \]  

(1)

Where, \( T(I) \) is the transmission, \( \Delta T \) is the modulation depth, \( I \) is the incident peak power intensity, \( I_{sat} \) is the saturable power density, and \( T_{ns} \) is the nonsaturable loss. According to the measured data, the modulation depth, and the nonsaturable loss were calculated to be 6.2% and 20%, as shown in inset of Fig. 2(a) [region1]. Additionally, the \( I_{sat} \) was about 28 MW/cm², corresponding to incident peak power (Pin) of ~16.3 W.

As is known, microfiber in itself is a good nonlinear waveguide. Meanwhile, Bi₂Se₃ nanoplatelets has been demonstrated with giant nonlinear refractive index \((n_2 \approx 10^{-14})\). It is interesting to explore whether the nonlinear effect can impact on the mode-locking behavior of microfiber-based TI film SA device. A direct approach is to enhance the incident peak power and simultaneously test the output spectrum. In experiment, clearly evolution stages was observed in Fig. 2, which was quite different from the common evolution trend as reported in [49, 65]. It was noted that the transmission would be reversed beyond the peak power intensity of ~400 MW/cm² (corresponding to ~232 W of Pin) in region 2. However, the transmission would be increased after the peak power intensity of ~960 MW/cm² (corresponding to ~558 W of Pin), then decreased again when the peak power intensity was beyond ~1900 MW/cm² (corresponding to ~1.1 kW of Pin), as illustrated in region 3&4 in Fig. 2(a). Figure 2(b) shows the measured spectrum from 960 nm to 1700 nm at Pin of ~350 W. New spectral components at A (~1300 nm), B (~1190 nm) and C (1000~1140 nm), were observed from the spectrum. It evidenced that the nonlinear effect caused the transition in the transmission curve. The corresponding mechanism maybe related with the four-wave-mixing (FWM) effect or Raman effect [50], which still need further investigation.

To this extend, we could give a brief predict on the possible operation behaviors depending on the peak power \( P_{in} \) in laser cavity. In region 1, stable soliton state could be maintained below \( P_{in} = \sim 232 \) W. As a contrast, the prerequisite for mode-locking no longer existed in region 2 due to the existence of nonlinear effect. However, the nonlinear effect played a curial role for the HML generation from the other point of view: once the peak power of soliton pulses reached to the \( P_{in} = \sim 232 \) W, further increasing of the pump power would force the pulse to split to lower their peak power for stable soliton operation. Larger pump power attributed to higher-repetition soliton pulse generation. But some other type of pulse could exist including of noise pulse and broadband-spectrum pulse [66].

3. Passive HML fiber laser setup

Figure 3 showed the schematic of mode-locked fiber laser with our TISA device. The pump source was a laser diode (LD) with emission centered at 976 nm. A piece of 2.4 m-long EDF
was used as the laser gain medium with absorption coefficient of 25 dB/m@980 nm (IsoGainTM I-25, Fibercore). The pump was delivered into EDF via a 980/1550 fused wavelength division multiplexer (WDM) coupler. A polarization dependent isolator (ISO), placed after the EDF, was used to ensure unidirectional operation and eliminate undesired feedback from the output end facet. A fused fiber optical coupler (OC) was used to extract 30% energy from the cavity. A polarization controller (PC), consisting of three spools of SMF-28 fiber, was placed in the ring cavity after the ISO. To depict the orientation of PC, the rotation angle $\Phi$ was set according to the laser direction in cavity. The TI-based SA was inserted between the PC and the WDM coupler. Apart from the gain fiber, all the fiber devices in cavity were SMF-28 fiber. The total cavity length was ~12 m. It should be pointed that the net dispersion of total cavity was difficult to be determined because of the following two reasons: 1) The dispersion coefficient of this type of EDF was unavailable on the web of Fibercore.com. 2) Our SA was a hybrid structure composed by the inner microfiber and the outer coated thin TI film, the dispersion of which could not be accurately valued. Firstly because that the waveguide dispersion of microfiber was changed along its taper region. Secondly, because the coated TI film was a high third-order nonlinear optical material [34], its impact on the light in microfiber still needed a further investigation when we considered its varied thickness around the microfiber surface. The performance of the laser was observed using an optical spectrum analyzer (Yokogawa, AQ6370B), 1 GHz digital oscilloscope (Tektronix, DPO7104C), 3 GHz RF spectrum analyzer (Agilent, N9320A) coupled with a 15 GHz photodetector (EOT, ET-3500FEXT), and an optical autocorrelator (APE, PulseCheck).

4. Results and discussion

4.1 FML performance

Continuous wave operation occurred at a pump power of ~32 mW, and the self-started mode-locking state appeared when the pump power was increased to ~35 mW. Herein, the rotation angle of three spools were settled to be $\Phi_1 = 40$ degree, $\Phi_2 = 180$ degree and $\Phi_3 = 0$ degree. The stable operation in FML was observed up to 51 mW of pump power. Figure 4(a) showed the typical spectrum of mode-locked pulses at the pump power of 44 mW. The generated optical soliton spectrum was centered at 1562.4 nm with a 3 dB bandwidth of 8.2 nm. The evident symmetric Kelly sidebands on the spectrum indicated that the mode locked laser was operating in the soliton regime. The first-order symmetric Kelly sidebands located at 1550.8 nm and 1574 nm. The measured autocorrelation trace of the output pulse was presented in the inset. The pulse duration $T_{\text{pulse}}$ was 320 fs if a sech$^2$ pulse profile was assumed. Correspondingly, the time-bandwidth product (TBP) of the pulses was about 0.323. The fundamental repetition rate was 17.34 MHz, which was determined by the ~12 m cavity length. The corresponding radio frequency (RF) spectrum of the laser was shown Fig. 4(b).
The SMSR was 75 dB measured with 20 kHz resolution bandwidth (RBW). The RF spectrum was quit flat for the 1 GHz scanning range with an RBW of 20 kHz. In this case, the FML state was extremely stable in the whole experiment. The average output power was around 0.56 mW. Therefore, the pulse energy was 32 pJ.

Herein, the cavity dispersion parameter $\beta_2$ can be evaluated by the well-known relation between the $m$th-order Kelly sideband position and the central wavelength in [45,67]. The $\beta_2$ could be easily deduced with a value of $-1.126 \times 10^{-2}$ ps$^2$/m. Considering that the cavity length was ~12m, the net cavity dispersion was estimated to be $-0.1351$ ps$^2$.

Fig. 4. Performance of FML state. (a) Spectrum and autocorrelation trace, (b) RF spectrum.

4.2 HML performance

The pulse fission was observed when pump power exceeds 51 mW. After slightly adjustment of the intracavity polarization state, the pulse interval and peak power could be equalized and the laser started to operate in HML regime. Then keeping the PC orientation fixed and further increasing the pump power, the harmonic number (N) of fundamental soliton pulses could be increased gradually as shown in Fig. 5. Noteworthy, the HML pulse would undergo short time instability at each new pump level until new harmonic number of mode-locking state completely established owing to the self-arrangement phenomenon [52]. In each HML case, it stably operated for a few hours in our experiment. The videos at different frequency, including of 86.3 MHz (5th @66 mW, Media 1), 523 MHz (30th @200 mW, Media 2), 800 MHz (46th @260 mW, Media 3) and 1.55 GHz (89th @395 mW, Media 4), were presented at their corresponding pump powers. Due to the limited bandwidth of the 1 GHz digital oscilloscope, the pulse trace demonstrated a certain degree of distortion in the video as the repetition rate beyond 800 MHz, which was very common in the previous reports [49], [60]. As the pump power was increased to the maximum 395 mW, the output power was reached to 41.4 mW.
The laser performance at HML of 1.55 GHz was presented in Fig. 6. The output spectrum in Fig. 6(a) centered at 1563.7 nm with a 3 dB bandwidth of 5.6 nm, and the first-order symmetric Kelly sidebands located at 1551.8 nm & 1575.6 nm, respectively. The pulse duration $T_{\text{pulse}}$ and TBP were measured to be 480 fs and 0.33. Figure 6(b) presented the RF spectrum over 3 GHz span with a RBW of 30 kHz. It should be pointed that the attenuation coefficient was automatically fixed to 20 dB by the RF spectrum analyzer, which did not affect the SMSR. In this case, the SMSR was 50 dB. The inset showed the RF spectrum measured at RBW of 1 kHz within 1 MHz span range. It can be seen that the background noise was suppressed to 65 dB from the RF peak. In this case, the pulse energy was 26.7 pJ.

Finally, we carefully rotated $\Phi_1$ but fixed $\Phi_2$ and $\Phi_3$ at the maximum pump power to measure the relation between repetition rate and polarization state. Amazingly, the harmonic number could be quickly increased by raising $\Phi_1$ from 40 degree to ~70 degree with a precision of ~2 degree/adjustment. In this process, the HML of 1.63 GHz (N: 94th), 1.68 GHz (N: 97th), 1.72 (N: 99th), 1.98 GHz (N: 114th), 2.23 GHz (N: ~129th), 2.53 GHz (N: 146th), 2.78 GHz (N: 161th) and 2.95 GHz (N: 170th) were obtained. Further raising $\Phi_1$, the laser started to operate in the continous wave regime. This process was repeatable when we turned $\Phi_1$ in the range. Figures 7(a) and 7(b) and Figs. 7(c) and 7(d) presented the laser performance at 2.23 GHz and 2.95 GHz, respectively. In case of 2.23 GHz, the output spectrum centered at 1564 nm with a 3 dB bandwidth of 4.55 nm, the pulse duration $T_{\text{pulse}}$ and TBP were 609 fs and 0.34, respectively. The output power was measured to be 43.5 mW. The RF spectrum had a
SMSR of 48 dB in the full range. In case of 2.95 GHz, the output spectrum centered at 1564.1 nm with a 3 dB bandwidth of 3.3 nm, the pulse duration $T_{\text{pulse}}$ and TBP were 920 fs and 0.37, respectively. The output power was measured to be 45.3 mW. The RF spectrum had a SMSR of 37 dB in the full range. In each case, the spectra from 960 nm to 1700 nm were recorded. It was found that the pump light was totally depleted; moreover there was no additional spectral component apart from the soliton spectra, indicating that the measurement on output power of laser was accurate. Obviously, our experimental results, such as the spectral properties, SMSR, and HML behaviors, critically depended on the performance of the microfiber-based TI film SA.

Given that the TI filmy thickness was not a constant around the microfiber and the taper waist region had unavoidable slightly irregularity, we tended to believe that this behavior originated from polarization-dependent properties of our SA. It might modify the property of nonlinear saturable transmission, which led to a quite different fundamental soliton condition of the laser cavity. However, it was not easy to precisely characterize the polarization-dependent properties of this in-line SA under our present condition.

Fig. 7. Laser performance at 2.23 GHz and 2.95 GHz. (a,b) Spectrum and autocorrelation trace for case of 2.23 GHz, RF spectrum with a span of 3 GHz and a SMSR at a level of 48 dB, (c,d) Spectrum and autocorrelation trace for case of 2.95 GHz, RF spectrum with a span of 3 GHz and a SMSR at a level of 37 dB.

Table 1 summarized the passive HML EDFL performance of this work with those reported based on NPR, CNT, graphene and other TI SAs. It was found that the HML based on NPR still had the highest repetition position (up to 22.2 GHz) but the power conversion efficiency was the lowest. The CNT/SPF type SA achieved the highest mode-locking with ~4.9 GHz repetition and >40 dB SMSR. Interestingly in the reported HML with TI SA, our work achieved highest 2.95 GHz repetition rate with pulse duration of 920 fs and a SMSR of 37 dB. Compared with the Bi$_2$Te$_3$ nanosheets/microfiber case [49], the output power of our laser was increased by 9 times, and the pulse duration was paved into the femtosecond
regime. We believed that a further optimization of the SA parameters (such as taper waist, TI filmy thickness) together with the cavity parameters (such as cavity length, pump power and output coupler) was essentially required for the substantial enhancement the HML performance.

Table 1. Passive HML EDFL performance of this work with others. * Not available.

| HML type               | Pump power (mW) | Output power (mW) | Tpulse (fs) | Repetition (GHz) | SMSR (dB) | Ref     |
|------------------------|-----------------|-------------------|-------------|------------------|-----------|---------|
| NPR                    | 2200            | 54                | 1000        | 3.079            | 25        | [57]    |
| NPR                    | 2000            | *                 | 1000        | 22.2             | 6.52      | 26      | [58]    |
| CNT/SPF               | 395             | 20                | *           | 4.915            | 40        | [59]    |
| Graphene              | 230             | 9.6               | 900         | 2.22             | 40        | [62]    |
| Sb₂Te₃/SPF            | 255             | 4.5               | 2200        | 0.304            | 55        | [63]    |
| Bi₂Te₃ nanosheets/microfiber | 126           | 5.02              | 2490        | 2.04             | *         | [49]    |
| Bi₂Te₃ PLD film/microfiber | 395           | 41.4              | 480         | 1.55             | 50        | This work |
|                        | 395             | 43.5              | 609         | 2.23             | 48        |         |
|                        | 395             | 45.3              | 920         | 2.95             | 37        |         |

5. Conclusions

In this paper, a kind of microfiber-based TI SA was firstly reported based on the PLD method. It was found that this SA had interesting nonlinear saturable property owing to the strong nonlinear effect induced by microfiber and TI film. Upon employing on this SA, we achieved stable fundamental mode-locking (FML) at central wavelength of 1562.4 nm with pulse duration as short as 320 fs. By adjustment the intracavity polarization state at the maximum pump power, we also obtained stable HML with repetition rate up to 2.95 GHz and output powers beyond 45 mW. According to our knowledge, for the reported TI-based passive HML lasers so far, the obtained 2.95 GHz is the highest repitition rate, the output power of 45.3 mW is the highest, and the pulse duration is fully paved into femtosecond regime. Our obtained results as well as previously presented by Z. Luo [49], [65] revealed that the microfiber-based TI film SA is a promising device for practical multi-GHz ultrashort pulses generation.

Acknowledgments

Supported by the NSFC (61275144@ 61308049), Natural science fund of Guangdong province (S2013010012235), the foundation for scientific and technical innovation in Higher Education of Guangdong (2013KJCX0161), the Improvement and Development Project of Shenzhen Key Lab (ZDSY20120612094924467), the Science and technology project of Shenzhen City (JCYJ20120613172042264, JCYJ20130329142040731, JCYJ20140318091413568, JCYJ20120613112423982).