High-quality 2-μm Q-switched pulsed solid-state lasers using spin-coating-coreduction approach synthesized Bi$_2$Te$_3$ topological insulators

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Received 15 December 2017; revised 2 February 2018; accepted 2 February 2018; posted 2 February 2018 (Doc. ID 315809); published 27 March 2018

In this paper, the fabrication process and characterization of Bi$_2$Te$_3$ topological insulators (TIs) synthesized by the spin-coating-coreduction approach (SCCA) is reported. With this approach, high-uniformity nano-crystalline TI saturable absorbers (TISAs) with large-area uniformity and controllable thickness are prepared. By employing these prepared TIs with different thicknesses as SAs in 2-μm solid-state Q-switched lasers, thickness-dependent output powers and pulse durations of the laser pulses are obtained, and the result also exhibits stability and reliability. The shortest pulse duration is as short as 233 ns, and the corresponding clock amplitude jitter is around 2.1%, which is the shortest pulse duration in TISA-based Q-switched 2-μm lasers to the best of our knowledge. Moreover, in comparison with the TISA synthesized by the ultrasound-assisted liquid phase exfoliation (UALPE) method, the experimental results show that lasers with SCCA synthesized TISAs have higher output powers, shorter pulse durations, and higher pulse peak powers. Our work suggests that the SCCA synthesized TISAs could be used as potential SAs in pulsed lasers.

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OCIS codes: (140.3540) Lasers, Q-switched; (160.4760) Optical properties.

https://doi.org/10.1364/PRJ.6.000314

1. INTRODUCTION

Nowadays, pulsed lasers emitting at the 2-μm wave band are gaining more and more attention for their wide application in the fields of medicine, interferometric sensing, coherent light detection and ranging, the manufacturing industry, as well as the pumping of optical parametric oscillators working at 3–5 or 8–12 μm [1–4]. Due to the advantages of flexibility, simplicity, compactness, and efficiency, passive Q-switching technique has been proved to be a favorable method to obtain laser sources from the visible to the middle-infrared (MIR) region [5–8]. With respect to the wavelength of 2 μm, various saturable absorbers (SAs), such as Cr:ZnSe [9], Pb-doped glass [10], InGaAs/GaAs [11], as well as carbon nanotubes (CNTs) [7] have been widely reported. These conventional SAs, however, have the problem of a narrow absorption band; in spite of that, those SAs have merits of flexible design of parameters, a large ratio of saturable to non-saturable losses, and a high damage threshold. For example, carbon nanotube (CNTs) have been widely investigated for their broad wavelength range, low cost, simple fabrication process, short recovery time (≈1 ps), and high-speed third-order optical nonlinearity [7,12]. The cluster-induced losses and sample clarity also severely restrict their applications [13]. In the last few years, two-dimensional (2D) layered materials employed as SAs in mode-locked or Q-switched lasers have been broadly investigated and reported for their large nonlinearity and unique electronic band structure [14–22]. Since single- or few-layered graphene SAs were successfully applied in many kinds of mode-locked or Q-switched laser system [14,15], other kinds of 2D materials, such as black phosphorus (BP) [16,17], transition-metal dichalcogenides (TMDs) [18–20], and topological insulators (TIs) [21–28], have also received plenty of attention owing to their unique optoelectronic properties. When it comes to TIs, as reported in Ref. [21], a TI is also a Dirac material with a narrow topological non-trivial energy gap (0.16–0.3 eV) at its bulk state and a gapless metallic surface state. Just like graphene, due to the Pauli-blocking effect, the
saturable absorption of TI could happen when the TI is excited by strong light with the single photon energy exceeding the TI’s bandgap. Benefiting from its advantages of narrow energy gap and gapless metallic state at its surface or edge, TI is expected to show an ultra-broad bandwidth of saturable absorption operation. Up to now, TIs have been employed as SAs in many mode-locked and Q-switched lasers from the visible to the near-infrared (NIR) range and show good performances [22–24]. With respect to a Q-switched 2-μm laser with TIs as SAs, several works on the TISA Q-switched 2-μm fiber lasers have been reported [25,26]; however, the reported pulse widths were more than 1 μs. Recently, our group has demonstrated a solid-state passively Q-switched 2-μm laser with a Bi2Te3-SA synthesized by ultrasound-assisted liquid phase exfoliation method (UALPE) [27], where the shortest pulse duration of 620 ns has been obtained. The 620 ns pulse duration was once the record of the pulse width for 2-μm Q-switched lasers with TIs as SAs. In this work, we have obtained 233 ns pulse width, which we think is the shortest one ever among the reported results of TISA Q-switched 2-μm lasers. To further improve performance, such as shortening the pulse duration for fulfilling requirement of applications, it is consequently expected to achieve narrower-pulse-duration 2-μm passively Q-switched lasers with TISAs of thickness or optical nonlinearity controllable 2D materials.

Recently, the spin coating-coreduction approach (SCCA) was discussed and reported for synthesizing Bi2Te3 SAs and pulsing Q-switched laser at 1 μm [33]. The SCCA can directly grow crystalline Bi2Te3 on a sapphire substrate without the transfer procedure of TI nanoflakes. The TISAs prepared through this method have consistency over a large area and a controllable thickness. In this work, the SCCA synthesized TISAs were used as SAs in a 2-μm Q-switched laser. For comparison, a passively Q-switched laser with a UALPE method synthesized Bi2Te3 SA is also investigated [27]. The Q-switched laser with SCCA synthesized TISAs can generate pulses with higher average output powers, shorter pulse durations, and higher pulse peak powers in comparison with the Q-switched laser with UALPE synthesized TISAs. This indicates that the SCCA synthesized TISAs have huge potential applications in high-quality Q-switched lasers.

2. PREPARATION AND CHARACTERIZATION OF Bi2Te3 TOPOLOGICAL INSULATORS

The process of SCCA used to prepare Bi2Te3 films is the same as the work reported in Ref. [34]. First, Bi(NO3)3·5H2O and TeO2 were dissolved in propylene glycol under stirring with a small amount of HNO3 to obtain a transparent precursor mixture. To date, plenty of methods for preparing TISAs, such as the molecular beam epitaxy (MBE) and vapor–liquid–solid methods (VLS) [28], hydrothermal intercalation exfoliation (HIE) [29], liquid phase exfoliation (LPE) [30], the polyol methods [31], and the solvothermal methods [32], have been reported. In spite of the proved functionality of pulsing lasers with these methods, there are various drawbacks for each method. For example, the MBE and VLS are expensive and time-consuming. In HIE and LPE, it is hard to control the size of exfoliated nanostructure TIs and their samples’ quality is limited by the bulk TI materials. As for the polyol and solvothermal methods, the aggregation of TI nanoplates depresses their application in pulsed lasers.

Fig. 1. AFM images and height variations of four TISAs. (a) TISA1; (b) TISA2; (c) TISA3; (d) TISA4.
3. **Q-SWITCH OPERATION**

To further investigate the saturable absorption properties of the as-prepared Bi$_2$Te$_3$ samples and their application feasibility for pulsed solid state lasers at 2 μm, a simple linear cavity solid-state laser test was used. The schematic experimental setup is shown in Fig. 3. A commercial fiber-coupled diode laser emitting at wavelength of 790 nm with a maximum output power of 50 W was used as the pump source. The fiber core with numerical aperture (NA) of 0.22 was 100 μm in diameter, and a 1:1 imaging module was employed to focus the pump light into the Tm:LuAG crystal. A 4 mm x 4 mm x 8 mm Czochralski-technique-grown Tm:LuAG coated with anti-reflection (AR) from 750 to 850 nm (reflectivity < 2%) and 1930 to 2230 nm (reflectivity < 0.8%) on both end sides was used as a laser crystal, and the Tm$^{3+}$ ions’ doping concentration was 6%. The crystal was wrapped with indium foils and held in a brass heat sink for efficiently cooling the laser crystal during pumping, and the temperature was held at 16 °C with a water cooler. As shown in Fig. 3, a mirror M$_1$ (r = 200 mm) was employed as the input mirror coated with high-reflection (HR) coating from 1850 to 2100 nm (reflectivity > 99.9%) and AR coating from 750 to 850 nm (reflectivity < 2%). Besides, a plane mirror M$_2$ was employed as output coupler (OC). For comparison, three OCs with different transmittances of 2%, 3%, and 5% from 1820 to 2100 nm were used. In this work, one UALPE synthesized TISA sample [27] and three SCCA synthesized TISAs were employed as the Q-switch SAs. The distance between M$_1$ and M$_2$ was about 20 mm. A digital oscilloscope (1 GHz bandwidth, Tektronix DPO 7102, USA) and a fast InGaAs photodetector with a rising time of 35 ps (EOT, ET-5000, USA) were used to measure the pulse characteristics of the laser. The spectrometer (resolution 0.4 nm, APE GmbH, Germany) and the power meter (Coherent Inc., USA) were employed to measure the laser spectrum and yielded average output power, respectively.

Figure 4 shows the average output power of Q-switched lasers with TISAs with respect to incident pump power and transmittances of OCs. Clearly, the thickness-dependent output can be seen with TISA1, TISA2, TISA3, and TISA4 for OCs with different transmittances of 2%, 3%, and 5%, respectively. The highest output powers for TISAs at the same incident pump power were obtained from the cavity with 5% transmittance of OC. In fact, as shown in Refs. [38,39], the output energy of passively Q-switched laser can be optimized by selecting an appropriate output coupler when the incident pump power, the gain medium, and the saturable absorber are given. Hence, the output power may be higher if the output coupler with transmittance more than 5% is employed. However, usually, to some extent, with the increase of output coupler transmittance, the Q-switched laser pulse will become

![Fig. 2. Nonlinear transmittance curves versus input pulse influences.](image)

![Fig. 3. Experimental setup of the Q-switched laser.](image)
relatively short pulse widths, we chose three mirrors with relatively low transmittance. In addition, the power of Q-switched lasers with TISA4 reveals the reduction with increasing pump power further. Generally speaking, the thermal stability of saturable absorber is crucial for passively Q-switched lasers with TISA4.

Therefore, in order to obtain Q-switched lasers with 5% transmittance of OC versus incident pump powers are then analyzed and shown in Figs. 5(a) and 5(b). All are with clear pump power dependency. The highest power, highest repetition rate, and shortest pulse duration of the Q-switched laser using TISA4 as saturable absorber are close to the reported results and around 1.25 W, 98.5 kHz, and 671.2 ns, respectively [27]. In comparison to the laser using TISA4, however, the pulse durations and repetition rates using TISA1, TISA2, and TISA3 are with even better performance and clear thickness dependency. First, using incident pump power of 9.81 W, the shortest pulse durations for Q-switched lasers with TISA1, TISA2, and TISA3 are 435.4, 389.5, and 233.3 ns, respectively. The inset of Fig. 5(a) is the temporal profile of the shortest duration of the laser with TISA3 under pump power of 9.81 W. The decrease of the pulse duration could be attributed to higher modulation depth in the thicker TISAs [34]. Second, the laser pulse repetition rates become higher with increasing thickness of the TISAs. For example, the corresponding highest pulse repetition rates of TISA1, TISA2, and TISA3 are 105, 122, and 145.5 kHz, respectively, as shown in Fig. 5(b). These results all indicate that the SCCA synthesized TIs can be used as SAs with controllable nonlinear parameters for a pulsed laser.

Here, one should note that SCCA-based TISAs have an advantage of uniformity. As shown in Fig. 1, TISA4, grown by the UALPE approach, exhibits clearly uneven distribution, leading to extra thermal loss due to poor thermal dissipation efficiency as pumping goes higher. This can be used to explain why the output power of the Q-switched laser with TISA4 decreases "over-saturation" due to the low saturation intensity of the TISA. At the state of "over-saturation," the accumulated heat on the TI sample causes an extra loss that makes the Q-switching laser performance deteriorated; hence, the output power will decrease [24]. However, even at the state of "over-saturation," we did not find damage to the TISAs. This is because the power intensity on SCCA-TISA in the cavity is about dozens of kilowatts per centimeter squared (kW/cm²) in our experiment, while as reported in Ref. [41], at a power intensity of 26.7 MW/cm², the Bi₂Te₃-SA can still work without any damage. This indicates that the TISAs have a damage threshold larger than tens of megawatts per centimeter squared (MW/cm²) and the related questions will be further investigated in future work.

In addition, the pulse durations and the repetition rates of these Q-switched lasers with 5% transmittance of OC versus incident pump powers are then analyzed and shown in Figs. 5(a) and 5(b). All are with clear pump power dependency. The highest power, highest repetition rate, and shortest pulse duration of the Q-switched laser using TISA4 as saturable absorber are close to the reported results and around 1.25 W, 98.5 kHz, and 671.2 ns, respectively [27]. In comparison to the laser using TISA4, however, the pulse durations and repetition rates using TISA1, TISA2, and TISA3 are with even better performance and clear thickness dependency. First, using incident pump power of 9.81 W, the shortest pulse durations for Q-switched lasers with TISA1, TISA2, and TISA3 are 435.4, 389.5, and 233.3 ns, respectively. The inset of Fig. 5(a) is the temporal profile of the shortest duration of the laser with TISA3 under pump power of 9.81 W. The decrease of the pulse duration could be attributed to higher modulation depth in the thicker TISAs [34]. Second, the laser pulse repetition rates become higher with increasing thickness of the TISAs. For example, the corresponding highest pulse repetition rates of TISA1, TISA2, and TISA3 are 105, 122, and 145.5 kHz, respectively, as shown in Fig. 5(b). These results all indicate that the SCCA synthesized TIs can be used as SAs with controllable nonlinear parameters for a pulsed laser.

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![Fig. 4](image_url) Average output powers of Q-switched lasers with different TISAs and different OCs.

![Fig. 5](image_url) Laser performances of Q-switched lasers with different TISAs versus incident pump powers. (a) Pulse durations; inset shows the temporal profile of the shortest duration of laser with TISA3 under pump power of 9.81 W; (b) pulse repetition rates; (c) single pulse energies; (d) pulse peak powers.
when the incident pump power exceeds 7 W. Moreover, the pulse duration cannot be further depressed with increasing pump power. In contrast to TISA4, using SCCA-based TISAs, the output power increases monotonically with the increasing pump power. The pulse durations also decline accordingly with the increasing pump power. For example, as shown in Fig. 1(c), the SCCA-based TISA3 has more uniform TI flake distribution. Hence, its thermal dissipation efficiency is better than that of TISA4, and the problem of extra thermally induced loss is also relatively slight. Therefore, the SCCA-based TISA3 has better output power performance and shorter pulse width than TISA4. These all reveal SCCA-based TISAs have better potential for high power applications.

Furthermore, the single pulse energies and pulse peak powers can be estimated according to Eqs. (1) and (2),

\[ E = P/f, \]  
\[ P_p = E/\tau, \]

where \( E, P, f, P_p, \) and \( \tau \) are the single pulse energy, the average output power, the pulse repetition rate, the peak power, and the pulse duration of the output laser, respectively. Figures 5(c) and 5(d) give the estimated pulse energies and the peak powers of Q-switched lasers with TISAs versus incident pump powers. The highest peak powers for TISAs from TISA1 to TISA4 are 35.6, 31.5, 50.5, and 24.1 W, respectively. For SCCA synthesized TISAs, the pulse peak power of laser with TISA3 is the highest among these TISA Q-switched lasers while the single pulse energy for a laser with TISA1 is higher than those of lasers with TISA2 and TISA3. This is because the laser with TISA3 has the shortest pulse duration.

In order to demonstrate the temporal quality of the pulses, the pulse train was characterized. Figure 6(a) shows one example of pulse train of laser with TISA3 as the absorber under a pump power of 9.81 W. The repetition rate is 145.5 kHz, and the pulse train is comparatively stable. We then evaluated the quality of pulse–amplitude equalization. This can be characterized by clock amplitude jitter (CAJ), which is defined as the ratio of the standard deviation (\( \sigma \)) to the mean value (\( M \)) of the intensity histogram at the pulse peak intensity, as in the following formula [29]:

\[ \text{CAJ} = \frac{\sigma}{M} \times 100\%. \]  

The CAJ of the pulse train with the highest pulse energy (11.8 \( \mu J \)) was calculated to be 2.1%, revealing good intensity stability. The inset of Fig. 6(a) is the corresponding optical spectrum and shows a clear emission wavelength of 2021.7 nm. In comparison with Ref. [27], there is a 3-dB optical bandwidth of the output pulse at the maximum power. This results from the introduced insertion losses induced by the SAs, in which the wavelength of a quasi-three energy level laser system will easily shift when switching the laser operation regime from continuous wave (CW) to Q-switching. Thus, if an extra element is employed in the cavity to vary the insertion loss, the central wavelength of the Q-switched laser would be tuned. In order to verify the sustainability of our laser system, the stability of average output power and pulse duration for a Q-switched laser with TISA3 at the incident pump power of 9.81 W was measured. We took the values every 5 min, and the whole measurement process lasted 2 h, as seen in Fig. 6(b). The average output power is around 1.74 W and with variation of about 10 mW. Clearly, within 2 h, the output powers of the lasers are within 1% variation. These all indicate the pulse quality of lasers using SCCA synthesized TISAs. Moreover, we compare the published works of 2-\( \mu \)m passively Q-switched lasers using

<table>
<thead>
<tr>
<th>SA</th>
<th>Gain Medium</th>
<th>Wavelength</th>
<th>Output Power</th>
<th>Pump Efficiency</th>
<th>Pulse Width</th>
<th>Refs.</th>
</tr>
</thead>
<tbody>
<tr>
<td>BP fiber</td>
<td>Tm(^{3+}), Ho(^{3+}) fiber</td>
<td>1912 nm</td>
<td>71.7 mW</td>
<td>11.95%</td>
<td>731 ns</td>
<td>[16]</td>
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<tr>
<td>BP</td>
<td>Tm:YAP</td>
<td>1988 nm</td>
<td>151 mW</td>
<td>4.47%*</td>
<td>1.78 ( \mu )s</td>
<td>[17]</td>
</tr>
<tr>
<td>BP</td>
<td>Tm:YAP</td>
<td>1969 nm and 1979 nm</td>
<td>3.1 W</td>
<td>41.2%*</td>
<td>181 ns</td>
<td>[42]</td>
</tr>
<tr>
<td>BP ceramic</td>
<td>Tm:YAG</td>
<td>2009 nm</td>
<td>38.5 mW</td>
<td>0.5%*</td>
<td>2.9 ( \mu )s</td>
<td>[43]</td>
</tr>
<tr>
<td>MoS(_2)</td>
<td>Tm, Ho:YGG</td>
<td>2085 nm</td>
<td>206 mW</td>
<td>1.7%</td>
<td>410 ns</td>
<td>[19]</td>
</tr>
<tr>
<td>MoS(_2)</td>
<td>Tm:GdVO(_4)</td>
<td>1902 nm</td>
<td>100 mW</td>
<td>3.2%*</td>
<td>800 ns</td>
<td>[20]</td>
</tr>
<tr>
<td>MoS(_2)</td>
<td>Tm:CLNGG</td>
<td>1979 nm</td>
<td>62 mW</td>
<td>2.4%*</td>
<td>4.84 ( \mu )s</td>
<td>[44]</td>
</tr>
<tr>
<td>MoS(_2)</td>
<td>Tm, Ho:YAP</td>
<td>2129 nm</td>
<td>399 mW</td>
<td>4.7%</td>
<td>435 ns</td>
<td>[45]</td>
</tr>
<tr>
<td>WS(_2)</td>
<td>Tm:LuAG</td>
<td>2013 nm</td>
<td>1.08 W</td>
<td>15.4%</td>
<td>660 ns</td>
<td>[46]</td>
</tr>
<tr>
<td>Bi(_2)Te(_3)</td>
<td>Tm(^{3+}), Ho(^{3+}) fiber</td>
<td>1893 nm</td>
<td>0.68 mW</td>
<td>0.27%</td>
<td>1.71 ( \mu )s</td>
<td>[25]</td>
</tr>
<tr>
<td>Bi(_2)Te(_3) fiber</td>
<td>Tm(^{3+}) fiber</td>
<td>1957.6 nm</td>
<td>26 mW</td>
<td>3.3%</td>
<td>2.22 ( \mu )s</td>
<td>[26]</td>
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<tr>
<td>Bi(_2)Te(_3)</td>
<td>Tm:LuAG</td>
<td>2023.6 nm</td>
<td>2.03 W</td>
<td>16.9%</td>
<td>620 ns</td>
<td>[27]</td>
</tr>
<tr>
<td>Bi(_2)Te(_3)</td>
<td>Tm:LuAG</td>
<td>2021.7 nm</td>
<td>1.74 W</td>
<td>17.7%</td>
<td>233 ns</td>
<td>This work</td>
</tr>
</tbody>
</table>

*The pump efficiency value with "*" at the top right corner represents the "output power/absorbed pump power," while the value without "*" represents the "output power/incident pump power."
2D materials as saturable absorbers, including BP, MoS2, WS2, and Bi2Te3. Table 1 shows the list of reported work about 2-μm Q-switched lasers. Except for Ref. [42], our work performs the shortest pulse duration and has relatively high output power among these Q-switched laser sources. One should note that the sustainability of the work of Ref. [42], which demonstrated a dual-wavelength laser with the highest output power of 3.1 W and the shortest pulse duration of 181 ns, is suspect due to BP materials' instability in air [47]. From the daily test of the pulse laser performance, the characteristics of the SCCA synthesized TISA-based pulsed laser are, however, with nearly fixed values under the same pump power. This also reveals the robustness of the SCCA synthesized TISA stored in the air. Therefore, we believe that the SCCA synthesized TISAs can be deemed as promising SAs in 2-μm solid-state pulsed lasers.

Previously, the difference of n and p type Bi2Te3 saturable absorption has been investigated [23]. By adjusting the density of the upper level for occupying as excitation through tuning the Fermi level, nonlinearity engineering was proposed for the pulsed laser. Unlike a typical semiconductor, however, it is not trivial for a topological insulator since the preparation of n or p type is difficult. Here, using the SCCA synthesized TISAs, the nonlinearity could be managed through adjusting the thickness of the TISAs resulting from dynamical modulation between bulk and surface state [34]. Besides, due to the Tl's nature of broadband absorption, stable and reliable Q-switched solid-state lasers beyond 1 μm and 2 μm are also expectable. Moreover, the large area uniformity of the SCCA synthesized TISAs also makes the scaling from NIR to MIR Q-switched solid-state lasers possible due to their nature of high thermal conductivity and optical nonlinearity tenability.

4. CONCLUSION

In summary, by employing the SCCA method, scaling a 2-μm passively Q-switched solid-state laser with stable output was reported. This is due to the unique nature of the SCCA synthesized TISAs, which are of high-uniformity nano-crystalline within a large area and of controllable thickness. The laser performances of 2-μm solid-state passively Q-switched lasers with SCCA synthesized TISAs with different thicknesses are measured. At an incident pump power of 9.81 W, the shortest pulse duration of 233 ns is obtained, and the corresponding clock amplitude jitter is around 2.1%. As far as we know, this is the shortest pulse duration in a 2-μm passively Q-switched laser with a TISA. The most important issue is that scaling laser power becomes possible due to the better thermal conductivity of TISAs resulting from the better uniformity. Consequently, in comparison with the TISA synthesized by the UALPE method, the lasers with SCCA synthesized TISAs have higher output powers, shorter pulse durations, and higher pulse peak powers. All the experimental results indicate that the SCCA synthesized TISAs could be used as potential SAs in pulsed lasers.

Funding. National Natural Science Foundation of China (NSFC) (61775119, 61378022, 61475088, 61605100); Young Scholars Program of Shandong University (SDU) (2015WLJH38); Ministry of Science and Technology, Taiwan, China (MOST 106-2112-M-110-006-MY3).

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