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Abstract: The successful demonstration of Q-switched and mode-locked fiber-laser operations using a topological insulator (TI) as saturable absorber (SA) has opened an application window besides TI’s originally expected features. However, to date, all-solid-state mode-locked lasers base on TISAs are still unavailable and became a desired goal not only due to their application as light sources, but also because of providing a way for deeper investigation of the nature of ultrafast dynamics present in TISAs. In this paper, the realization of a continuous-wave mode-locked all-solid-state laser with a repetition rate of around 1 GHz is reported using a high-quality TI SA mirror (TI-SAM) with ultralow saturation intensity, fabricated by a spin coating-co-reduction approach. An output power of 180 mW and pulse duration of 8 ps are observed. In addition, a 61 dB pulse-train quality from the radio frequency spectrum of mode-locked operation prove the feasibility of the proposed laser. To the best of our knowledge, this is the first experimental demonstration of a mode-locked solid-state laser based on TIs. In addition, this paper shows that the use of TISAs is a promising option for the realization of scaling solid-state mode-locked lasers with higher repetition rates, reaching order of GHz.

Index Terms: Topological insulator (TI), saturable absorber (SA) mirror, mode-locked laser.

1. Introduction

Ultrafast lasers are a topic of focus for quantum electronics due to their wide-ranging military, scientific, medical, and industrial applications [1]. Since the 1960s, numerous approaches have been proposed and successfully demonstrated for constructing mode-locked lasers [1]. Benefiting from the compactness and low cost, passively mode-locked lasers based on saturable absorbers (SAs) have led to such lasers becoming important tools for generating ultrafast laser pulses with pulse...
widths ranging from picoseconds to femtoseconds. Recently, multi-gigahertz ultrafast lasers have attracted increasing attention and been extensively studied because of the tremendous growth of their potential applications, such as optical frequency comb generation, Doppler-free spectroscopy, high-speed optical sampling, and optical communication [2]. Various methods have been proposed and demonstrated to realize such laser systems, such as harmonic mode-locked fiber lasers, semiconductor lasers, solid-state lasers, and so on [2]. Mode-locked solid-state lasers exhibiting high power, broad bandwidth, and a repetition rate ranging from several GHz to 160 GHz have been already reported [3]–[6]. It should be emphasized that a compact high-repetition-rate mode-locked solid-state laser requires a sophisticated laser cavity configuration, and the single-pulse energy will be reduced accordingly. Moreover, to achieve the saturation conditions, cavity design using a tightly focused SA is unavoidable. This leads to increased cavity complexity and SA damage [2]. Therefore, new saturable-absorption materials exhibiting low saturation intensity are critical and have become an important research target for ultrafast optoelectronics.

Recently, quantum-confined materials, such as quantum dots or metallic nanoparticles, have been observed to possess low saturation intensity due to their limited density of states in the upper levels occupied by excited electrons [6], [7]. However, their difficulty of preparation and poor uniformity limit their applicability. Fortunately, two-dimensional (2D) materials provided remarkable progress in the application of SAs for creating pulsed lasers [8]. Not only Q-switched, but also mode-locked operation has been achieved using 2D materials, such as graphene [9], topological insulators (TIs) [10]–[12], transition-metal dichalcogenides (TMDs) [13], [14], and black phosphorus (BP) [15], [16]. Using these materials, mode-locked lasers with a repetition rate of few GHz have also been reported [17]. For example: In 2012, Bernard et al. first verified the saturable absorption property of Bi$_2$Te$_3$ at 1.5 μm [18]. Zhao et al. reported their work about measuring the saturation properties of Bi$_2$Te$_3$ and Bi$_2$Se$_3$ and realizing a passively mode-locked fiber laser [10], [11]. Following that study, TIs have been successfully used for mode-locking or Q-switching fiber lasers operating at various wavelengths ranging from 0.6–3.0 μm [19]–[28]. In 2016, our group demonstrated a low-pulsing-threshold Q-switched mode-locked (QML) solid-state laser using a large area Bi$_2$Te$_3$-based TISA and investigated the saturable absorber sensitivity to the nature of the bulk state [29]. Besides, the reversible dynamic transition from Q-switched pulse mode to continuous-wave mode was observed experimentally on a topological insulator-based (Bi$_2$Te$_3$) passively Q-switched (QS) solid-state laser and explained by modulation-instability (MI) analysis based on the laser rate equations [30]. Base on these works, therefore, it can be noted that the role of the bulk state is crucial and its contribution during laser build-up cannot be neglected. In addition, this indicates that TISA might be a possible candidate for the construction of high-repetition-rate pulsed lasers due to its nature of low saturation intensity originating from its intrinsic bulk state. In this letter, we report the first realization of a CWML solid state laser using a TISA; the energy intensity incident onto the TISA is decreased using a large spot size on the SA in a compact short cavity. For fulfilling the uniformity requirements of SAs in the proposed cavity, a topological insulator saturable absorber mirror (TI-SAM) with high quality and large-area consistency was prepared by spin coating-co-reduction approach (SCCA). A maximum output power of 181 mW with a repetition rate of 949 MHz was obtained, corresponding to a slope efficiency of 36%. A clear transition from CW operation to CWML was observed and discussed. This study demonstrates a new method and window for the realization of a TISA-based mode-locked solid-state laser with a high repetition rate of up to ∼GHz.

2. TI-SAM Preparation and Characterization

TIs are unique phases of quantum matter and currently a highly focused topic in condensed-matter physics. Similar to graphene, TIs show a Dirac-like linear band dispersion for their surface states, which results from band conversion due to strong spin-orbital interaction. Hence, they attracted tremendous interest in various fields, such as spintronics, anomalous quantum Hall effect (AQHE), and so on [31]. In addition to the distinct band structure for the surface states, TIs also behave as intrinsic insulators with a narrow band-gap (0.16–0.3 eV) for the surface states separated from
the underlying bulk states. This narrow band-gap for TIs has been used for constructing a SA operating in a wide wavelength range. Currently, the common methods used to prepare TISAs involve hydrothermal intercalation exfoliation (HIE) [32]–[34], liquid phase exfoliation (LPE) [35], [36], polyol methods [37], and solvothermal methods [38], [39]. However, it is difficult to obtain good uniformity in TISAs prepared by these methods because of the inevitable clustering of the TI nanoplates solution deposited onto the substrate. This severely limits its applicability in solid-state ML bulk lasers, which require a large spot size on the SA. Pulsed laser deposition (PLD) is an established approach for depositing uniform TI films on substrates, but the equipment for PLD tends to be expensive [40], [41]. Recently, spin coating-co-reduction approach (SCCA) was proposed and demonstrated for directly growing crystalline Bi₂Te₃ on top of a sapphire substrate without the transfer motion of TI nanoflakes [42]. In this method, the oxide material solution is initially spin-coated and then co-reduced in a reducing atmosphere to synthesize c-axis oriented thin films. The TISAs prepared this way show consistency over a large area and a controllable thickness [43]. Previously, by tuning the thickness of the TISA, an adjustable optical nonlinearity of the TISA was identified and engineered into a pulsed solid-state laser with a large-area uniform SCCA grown TISA [42], [43]. In our experiments, SCCA was used to prepare large-area uniform TI films. A photograph and schematic of the TI-SAM are shown in Fig. 1(a) and Fig. 1(b), respectively. First, the Bi₂Te₃ films were grown on one side of a double-polished sapphire substrate with dimensions of 9 × 9 × 0.4 mm³ by SCCA. Then, a gold coating with a thickness of 200 nm was deposited onto the opposite side of the sapphire substrate by e-beam evaporation. After this metallization, the TI-SAM was successfully fabricated.

As shown in Fig. 2(a), the morphology of the TI-SAM was investigated by atomic force microscopy (AFM). Fig. 2(b) shows its thickness to be in the range of 6–12 nm, and its lateral dimensions in the range of 50–200 nm, corresponding to the solid line marked in Fig. 2(a). According to the statistical AFM analysis, the thickness distribution is obtained in an area of 25 μm² [Fig. 2(c)]. The major thickness of the TI-SAM is 8–10 nm, corresponding to 8–10 quintuple layers (QLs). The prepared TI-SAM was characterized by a Raman spectrometer excited by a 532 nm laser; the Raman spectrum is shown in Fig. 2(d). The data show three typical peaks at 62.3, 103.6, and 135.8 cm⁻¹, consistent with the A₁₁g, E₂g, and A₂₁g vibrational modes of Bi₂Te₃ films, respectively [44].

The saturable absorption behavior of the TI-SAM was measured by a femtosecond laser. Its nonlinear transmission curve is shown in Fig. 2(e). First, the reflectance of the sapphire-gold substrate without the Bi₂Te₃ film was measured to be 96.3%. Considering the measured substrate loss of 3.7%, the unsaturable loss of the TI-SAM was calculated to be 2.3%. Using the fitting formula in Ref [45], the modulation depth and saturation intensity were 8% and 210 W/cm², respectively. The saturation intensity was much lower than that found at the GW/cm² level in previous reports [10]–[12] but similar to reports at the ultralow level [30], [34], [38], [42], [43].
3. Passively Mode-Locked Laser

Typically, the focusing scheme used in lasers incorporating a saturable absorber within the laser cavity is designed based on the provision of a high intensity condition for fulfilling the saturation requirement. To saturate the SA, solid-state mode-locked lasers were widely used in Z-type or W-type cavities with a curved mirror to focus the beam onto the SA [13]. The cavity lengths were longer than 1 m, resulting in pulse repetition rates about 100 MHz [13]. However, owing to over-saturation or even transition from Q-switched to CW operation for the given low saturation intensity TISAs, a conventional cavity scheme seems unsuitable [29], [42], [43]. Therefore, developing a cavity for a pulsing solid-state laser using TISAs is an inevitable and critical issue. To realize a mode-locked laser based on a TI-SAM with ultralow saturation intensity, we designed a cavity with a decreased pulse intensity incident on the SA. The relatively shorter length of the laser cavity made increasing the pulse repetition rate possible. In addition, the spot size on the TI-SAM was designed to be several times larger than in previous studies to further reduce the beam energy density [13].

In this work, as shown in Fig. 3, a V-type cavity was used to realize a TI-based ML solid-state laser because the cavity mode in the linear cavity with plane mirror is hard to match the pump beam and a fold mirror is used as an output mirror. In addition, a folding V-shaped laser cavity can minimize the residual pump to the broadband saturable absorber [34]. A $4 \times 4 \times 7 \text{mm}^3$ Nd:YVO$_4$ crystal with 0.5% Nd$^{3+}$ doping concentration was directly pumped by an 808 nm fiber-coupled laser with a core diameter of 105 $\mu$m and a numerical aperture of 0.22. The pump beam was focused into the crystal with the beam waist of 59.5 $\mu$m $\times$ 60.6 $\mu$m using a 1:1 optical collimation system. The confocal parameters of the pump beam is 2.3 mm. The input concave mirror (IM $R = 100$ mm) was...
coated for high reflection at 1064 nm \((R \geq 99.9\%)\) and high transmission at 808 nm \((T \geq 95\%)\). A concave mirror \((R = 200\,\text{mm})\) with a transmittance of \(T = 10\%\) at 1064 nm was used as the output coupler (OC). The TI-SAM was mounted at the end of the cavity without active cooling. The length between IM and OC is 103 mm. The length between OC and TI-SAM is 48 mm. The laser pulse profile was recorded using a digital oscilloscope (Tektronix DPO7104, 1 GHz bandwidth) and a detector (New focus 1611, 1 GHz bandwidth). The output pulse duration and spectrum were monitored by an autocorrelator (APE, Pulse Check 150) and optical spectrum analyzer (Yokogawa AQ6370). The laser mode radii were calculated to be 60 and 500 \(\mu\text{m}\) in the laser crystal and on the TI-SAM, respectively, using ABCD matrix propagation theory.

Firstly, continuous-wave operation was realized with a high-reflection end mirror \((@1064\,\text{nm}\,R \geq 99.9\%)\) instead of TI-SAM. As shown in Fig. 4(a), the maximum average output power was 260 mW under an absorbed pump power of 630 mW, corresponding to a slope efficiency of 46\%. With the TI-SAM employed in the cavity, a QS laser was almost directly realized with a repetition rate of 163 kHz and pulse width of 2.26 \(\mu\text{s}\) when the absorbed pump power was around the threshold.
of 77 mW. The high repetition rate at low absorbed pump power can be attributed to the ultralow saturation intensity as has been discussed and reported previously [29], [34]. When the absorbed pump power was increased to 214 mW, the QML operation could be observed. Fig. 4(b) shows the repetition rate and pulse duration versus absorbed pump power. Clearly, as theoretically predicted, the repetition rate of the QS envelope increased accordingly with the increasing absorbed pump power. The most important observation was the CWML initiation and its operation with an absorbed pump power over 517 mW. This is the first time that CWML could be demonstrated using TIs as saturable absorber in diode pump all-solid-state lasers (DPSSL). Clear evolution of pulse train as increasing pump power was observed. Right above threshold, the laser ran in QS operation shown in Fig. 4(c). Further increasing pump power, higher pulse energy and shorter pulse duration enhances the modulation strength introduced by TI-SA. As a result it generated coherent frequency sideband separating by a free spectral range successively from the central frequency to startup ML with a QS envelope due to the QS instability, which is illustrated in Fig. 4(d). The Q-switching modulation frequency of QML pulse train was increasing as pump power which was revealed in Fig. 4(e) and QML consequently transitted to CWML shown in Fig. 4(f) when the pump power was further increased. It was due to the saturation of TI-SA that lead to the supression of QS instability. To further identify, output pulse trains of CWML with smaller time span was insert in the inset of Fig. 4(f). The maximum CWML output power was 181 mW under an absorbed pump power of 581 mW. A slope efficiency of 36% was estimated. When the absorbed pump power exceeded 600 mW, the laser was found to run into metastable regime. This could be due to the over saturation which is mainly caused by the low saturation intensity of TI-SAM. One should be noted that there is no optical damage or degradation observed during the experiments. The possible reason is the optical intensity will decrease tremendously as metastable starting. The intensity inside cavity can not be increased and the thermal accumulation will be therefore limited.

Temporal- and spectral-domain measurements were carried out to further confirm and investigate the characteristics of the pulses. Fig. 5(a) shows the autocorrelation trace of the mode-locked laser. The pulse duration was measured to be 7.9 ps by Gaussian fitting. The optical spectrum of QS and CWML is shown in Fig. 5(b). For QML operation, the spectrum is similar to the one of QS operation. The clear difference appears when QML operation transition to CWML as increasing the power, as shown in Fig. 5(b). The central wavelength of both the QS and CWML outputs is approximately 1064 nm. The spectra of CWML shows a clear broadening for the FWHM. Additionally, the radio frequency (RF) spectrum shows a clean fundamental peak at 948.9 MHz, which is consistent with the optical path of the cavity. The absence of parasitic side peaks and a signal-to-noise ratio (SNR) of over 61 dB confirm the mode-locked operation, as seen in Fig. 6. To further identify, the RF spectrum with smaller frequency span was insert in the inset of Fig. 6. The downturntrend observed in
the RF spectrum is mainly due to the spectral response of our photodetector [46]. In general, the RF spectrum was used to characterize the pulse quality. Compared to previous reported works about CWML using 2D materials, output of SCCA growth TISA based CWML shows the comparable mode locking quality. Furthermore, from the recorded FWHM of 0.24 nm, one can calculate the time bandwidth product to be approximately 0.502, which is 1.14 times the transform limit value of 0.44 for a Gaussian shaped pulse. The shorter pulse duration will be therefore expectable if the appropriate compensation of the dispersion inside cavity is employed. These data show and identify the quality of the pulsed-laser output.

Table 1

Comparative Characteristics for Ultrafast Gigahertz Lasers Based on Graphene, SESAM, and Ti

<table>
<thead>
<tr>
<th>Gain material</th>
<th>SA</th>
<th>P (mW)</th>
<th>Freq (GHz)</th>
<th>T (%)</th>
<th>(\omega) ((\mu)m)</th>
<th>E ((\mu)J/cm(^2))</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>Yb:KYW</td>
<td>SWCNT</td>
<td>47</td>
<td>1.2</td>
<td>0.3</td>
<td>30</td>
<td>302</td>
<td>[49]</td>
</tr>
<tr>
<td>Yb:KYW</td>
<td>SWCNT</td>
<td>35</td>
<td>1.13</td>
<td>0.3</td>
<td>30</td>
<td>239</td>
<td>[50]</td>
</tr>
<tr>
<td>VECSEL</td>
<td>Graphene</td>
<td>12.5</td>
<td>2.5</td>
<td>0.2</td>
<td>30</td>
<td>125</td>
<td>[17]</td>
</tr>
<tr>
<td>Yb:CALGO</td>
<td>SESAM</td>
<td>4100</td>
<td>5</td>
<td>2.6</td>
<td>100</td>
<td>100</td>
<td>[5]</td>
</tr>
<tr>
<td>Er:Yb:glass</td>
<td>SESAM</td>
<td>35</td>
<td>101</td>
<td>1.2</td>
<td>4</td>
<td>57.5</td>
<td>[51]</td>
</tr>
<tr>
<td>Nd:YVO(_4)</td>
<td>SESAM</td>
<td>45</td>
<td>157</td>
<td>0.2</td>
<td>18</td>
<td>14.1</td>
<td>[3]</td>
</tr>
<tr>
<td>Nd:YVO(_4)</td>
<td>MoS(_2)/Gra</td>
<td>404</td>
<td>0.94</td>
<td>10</td>
<td>128</td>
<td>8.4</td>
<td>[52]</td>
</tr>
<tr>
<td>VECSEL</td>
<td>QD-SESAM</td>
<td>100</td>
<td>50</td>
<td>1.6</td>
<td>60</td>
<td>1.1</td>
<td>[53]</td>
</tr>
<tr>
<td>Nd:YVO(_4)</td>
<td>Ti</td>
<td>181</td>
<td>0.95</td>
<td>10</td>
<td>500</td>
<td>0.13</td>
<td>This work</td>
</tr>
</tbody>
</table>

Note: P - output power; Freq - repetition rate; T - OC transmittance; \(\omega\) - beam waist on the SA; E - pulse fluence on the SA.
In the last five years, TISA-based mode-locked fiber lasers have been widely investigated. Although several studies have reported output powers for such lasers in the tens of mW range, their typical output powers are only a few mW [10], [11]. Moreover, the studies reporting higher powers mainly used a scheme in which the fiber sides are covered with TIs and operate at low repetition rates [40]. Additionally, GHz mode-locked fiber lasers have been reported using a harmonic mode-locking approach as well [20], [24], [47], [48]. One should be noted that pulse quality, fixed repetition rate and scalable power of all solid state laser are crucial for application. Therefore, our result shown exhibit the expectable future.

Table 1 shows a comparison of GHz mode-locked lasers based on graphene, SESAM, and TISAs. The pulse fluence on the TI-SAM was calculated to be approximately 0.1 \( \mu \)J. It is at least two orders of magnitude lower than that on graphene and SESAM. In addition, compared to mode-locked bulk lasers based on other 2D materials [18], the energy intensity incident on the TI-SAM is relatively low, yet still comparable to the saturation intensity of TI-SAMs. Unlike typical cavity designs of TISA-based solid-state lasers, the over-saturation due to the large intensity on the TISA is inevitable and results in unstable pulsing operation for further increasing pump power. Using SCCA to manufacture the TISA, a desirable large-area uniformity was achieved. This provided the necessary conditions for the cavity with the proposed focusing scheme. Moreover, the low saturable absorption of TISAs provide an opportunity for solving the problem of scaling power for high-repetition-rate pulsed lasers. It can be therefore noted that the low saturation intensity and large uniformity of SCCA TISAs provide a new way of constructing high-power and high repetition rate (\( \sim \)10 GHz) mode-locked solid-state lasers.

4. Conclusions

In conclusion, using high-quality large-area uniform SCCA grown TI-SAM, a \( \sim \)GHz repetition rate passively mode-locked solid-state mode-locked Nd:YVO\(_4\) laser was demonstrated for the first time to our best knowledge. Besides the two-hours-long operation, a signal-to-noise ratio (SNR) of over 61 dB in the RF spectrum shows the pulse quality of the mode-locked laser. Furthermore, an output power of \( \sim \)180 mW, with a single pulse energy of 0.1 nJ and pulse duration of \( \sim \)8 ps reveals the application potential. The success results from the suitable cavity design for the low saturation intensity of TISAs. Meanwhile, this work not only proves the role of the bulk state within the pulse build-up in the laser, but also provides a possible application of TIs in high-repetition-rate solid-state lasers.

References


