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Tm^{3+}-Doped Harmonic Dissipative Soliton Mode-Locked Fiber Laser at 1.93 μm Based on Tungsten Disulfide in Anomalous Dispersion Regime

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ABSTRACT We report a harmonic dissipative soliton 1.93 μm thulium fiber laser based on a tungsten disulfide (WS_2) saturable absorber (SA) in the anomalous dispersion regime. The multilayer WS_2 nanosheets were prepared by liquid phase exfoliation method and the SA was fabricated by dropping WS_2 solution onto a gold mirror. The transferred WS_2 SA had a modulation depth of 2.5% and a saturation intensity of 0.82 MW/cm^2. By incorporating the SA into a linear Tm^{3+} fiber laser cavity, harmonic mode-locked dissipative soliton laser was achieved at 1930 nm with the spectral width of 8 nm, the pulse energy of 3 nJ, the pulse width of 3.6 ns, and the repetition rate of 56.3 MHz. Based on the experimental results, it is shown that with the presence of harmonic mode-locking in 2 μm wavelength region, the multilayer WS_2 serving as a SA was verified to be a good candidate for broadband high-energy mode-locking. The order of the harmonic dissipative soliton mode-locked pulses remains the same along with the increasing pump power.

INDEX TERMS Mode-locked fiber laser, 2D material, tungsten disulfide, 2 μm fiber laser, Tm^{3+}-doped fiber.

I. INTRODUCTION

In recent years, passively mode-locked ultrafast lasers operating in the 2 μm region based on thulium-doped fibers have been intensively investigated for the important applications in remote sensing, medical surgery, industrial micromachining, and scientific experiments [1]–[4]. Look back to the historic evolutions of the mode-locked fiber lasers, it comes along with many types of mode-locked pulses including conventional solitons [5], [6], stretched pulses [7], [8], dispersion-managed soliton [9], [10], dissipative soliton (DS) [1], [11], [12], noise-like pulse [13], [14], and so forth. From the viewpoint of practical applications, high peak power and high pulse energy mode-locked fibers are always highly demanded. Since the pulse energy can be 1 $\sim$ 2 orders of magnitude larger than those from the conventional soliton mode-locking, DS has attracted considerable interests recently [13], [14]. Generally, most of the research works about DS were carried out in the 1 μm and 1.5 μm wavelength regions and only quite a few investigations for DS were proposed at 2 μm. This is because the dissipative solitons were exclusively found in all normal-dispersion mode-locked lasers theoretically [15]–[17]. Moreover, the commercially available gain fibers (GFs) in the 2 μm wavelength region owns relatively large anomalous dispersion, which results in 2 μm mode-locked lasing in the conventional soliton regime. However, the experimental investigations showed that DS can

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also be achieved in the mode-locked lasers operating in the anomalous dispersion [12], [18]–[20].

So far, to achieve passive mode-locking, there have been several approaches demonstrated in a Tm^{3+}-doped fiber laser (TDFL), such as nonlinear polarization evolution (NPE) [21], semiconductor saturable absorber mirrors (SESAMs) [22], [23], carbon nanotubes (CNTs) [24], and graphene [25]. However, there are some existing disadvantages for these methods. For example, the NPE suffers from the bulky and complicated configuration as well as the environmental instability. For SESAMs, the design for improving the damage threshold is complicated and the operating bandwidth is limited. As to the CNT-based saturable absorber (SA), the operating bandwidth is narrow since it is highly constrained by the average diameter of the nanotubes. The graphene also has two main disadvantages. One is the weak modulation depth (typically $\sim 0.4\%$ per layer at wavelengths about 2 μm [26]) and the other is the difficulty in creating an optical bandgap. Therefore, it is essential to explore new nonlinear optical materials for serving as broadband and cost-effective SAs with high damage threshold for 2 μm mode-locked fiber lasers.

More recently, the transition mental dichalcogenides (TMDs) such as Molybdenum disulfide (MoS_2) and Tungsten disulfide (WS_2) have received particular attentions due to their layer-dependent optoelectronic properties [16], [27]–[33]. Interestingly, it is well-known that the monolayered MoS_2 and WS_2 are direct bandgap materials whereas the multilayered MoS_2 and WS_2 are indirect bandgap materials. For example, the bandgap energy of monolayer WS_2 (direct bandgap) is $\sim$2 eV ($\sim$620 nm) [33], while the bandgap energy of bulk WS_2 (indirect bandgap) is $\sim$ 1.34 eV [29]. Accordingly, for the photons with the energy less than 1.34 eV, neither the monolayer WS_2 nor the bulk WS_2 can be applied as a SA device. However, the WS_2 has been experimentally demonstrated to exhibit saturable absorption property for the photons with the energy over the 0.62 - 1.24 eV range [11], [16], [30], [35]. It is found that the mechanism of generating the saturable absorption for WS_2 is deriving from the decreased defect-induced bandgap [16] or exciton effect [36]. Except for the existing saturable absorption effect, it is worth of noting that the layered WS_2 films also have a giant third-order optical nonlinear refractive index at the order of 10^{-13} m^2W^{-1} [36], [37], which is quite comparable to graphene (10^{-12} m^2W^{-1}) and is much larger than that of the carbon nanotubes (10^{-17} m^2W^{-1}). As a result, WS_2 can be a good option for playing as either a high nonlinear photonic device or the SA in the laser systems.

In this work, we demonstrate passive mode-locked harmonic dissipative soliton (HDS) in a TDFL based on a multilayered WS_2 SA. Normally, the HDS can be observed in the anomalous dispersion regime. However, to the best our knowledge, this is the first time that the HDS is demonstrated in TDFL at 1.93 μm operating in the anomalous dispersion regime. The repetition rate of the mode-locked TDFL was 56.25 MHz, which corresponds to the 3rd harmonics of the fundamental cavity frequency (18.75 MHz).

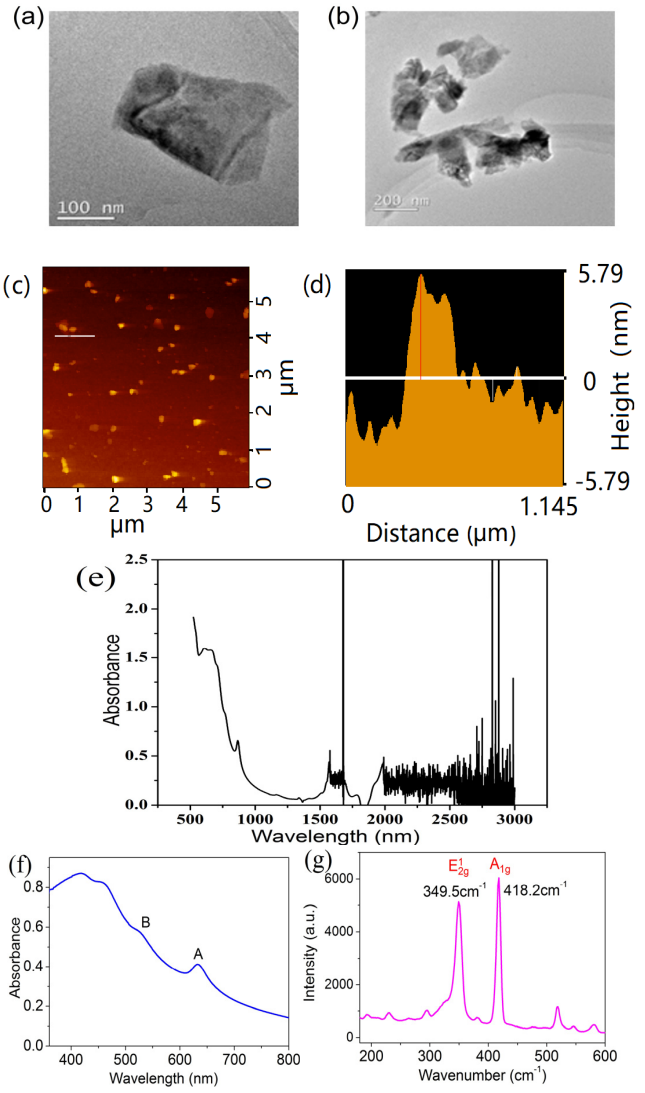

FIGURE 1. (a) and (b) TEM images for the WS_2 nanosheets under different magnification factors. (c) AFM image of the WS_2 nanosheets. (d) Measured thickness of WS_2 nanosheets using AFM. Absorption spectrum of the WS_2 dispersed liquid at (e) VIS-NIR-MIR and (f) visible spectral range. (g) Raman spectrum of the WS_2 dispersed liquid.

II. CHARACTERISTICS OF NONLINEAR MATERIAL

The multilayer WS_2 was synthesized through a liquid-phase exfoliation method (LPE) [37]. FIGURES 1(a) and 1(b) show the typical Transmission Electron Microscopy (TEM) images of the WS_2 dispersed liquid at different positions under different magnification factors. From the TEM images, it can reflect that the WS_2 are existing as 2D nanosheets in a liquid suspension form, which guarantees the successful exfoliation of the materials. The Atomic Force Microscopy (AFM) image of WS_2 nanosheets is also shown in FIGURE 1(c) and the average thickness is around 4 nm, as shown in FIGURE 1(d) [38], [39]. The measured VIS-NIR-MIR

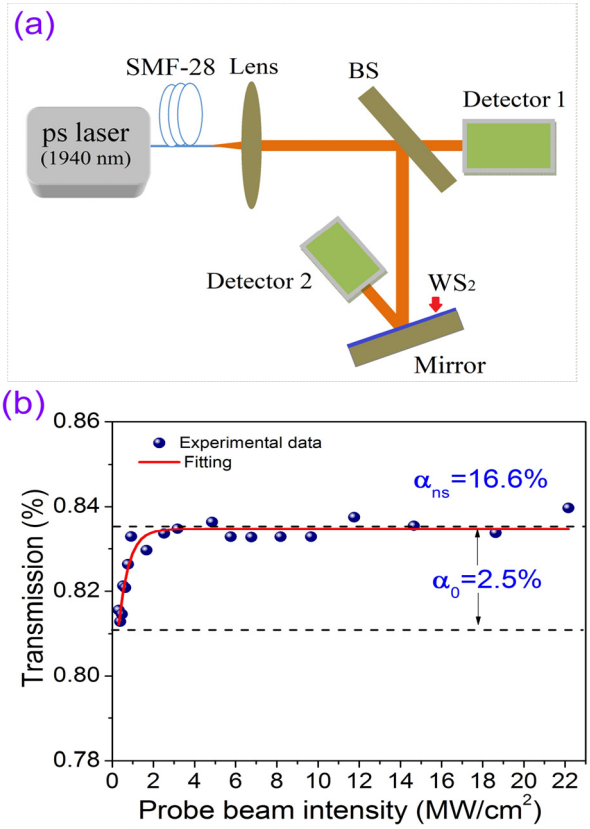

FIGURE 2. (a) Experimental setup for the nonlinear absorption characterization. (b) the measured saturable absorption characteristics of the WS_2 nanosheets coated on a gold mirror.

and UV-VIS absorption spectra of the WS_2 dispersed liquid are respectively shown in FIGURE 1(e) and 1(f). A strong absorption band can be found over the 1.93 μm band in FIGURE 1(e). From FIGURE 1(f), the presence of nanoscale WS_2 is supported by the observation of the excitonic absorption peaks A and B, around 630 and 550 nm, respectively. The absorption peaks are deriving from the direct bandgap transitions at the K point, corresponding to its "spin–orbital paired" absorption peaks [40]. The WS_2 nanosheets were further characterized by Raman spectroscopy using a 514 nm argon laser. As shown in FIGURE 1(g), two peaks located at 349.5 and 418.2 cm^{-1} are related to the in-plane (E^1_{2g}) and out-of-plane (A_{1g}) vibrational modes [36]. The wavenumber differences between the E^1_{2g} mode and A_{1g} mode is 68.7 cm^{-1}, which verifies that the WS_2 sample is multilayered.

In fabrication, tiny liquid solution full of dispersed WS_2 nanosheets was dropped onto a gold mirror and then dried with a heat gun to make few-layer WS_2 based SAs. Subsequently, a twin-detector measurement system was designed to investigate the nonlinear optical absorption characteristics of the WS_2-based SA, as shown in FIGURE 2(a). A 800 ps TDFL at 1940 nm with a repetition rate of 55 MHz was used as the pump source. The laser beam from this picosecond fiber laser was first collimated by a lens and was then divided

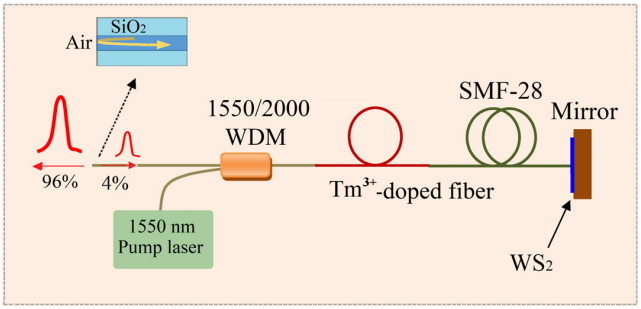

FIGURE 3. Experimental setup of the WS_2 nanosheet mode-locked Tm^{3+} fiber laser.

into two beams using a beam splitter (BS). One beam was used as the reference while the other beam was deflected to be incident on the WS_2 sample. After passing through the WS_2 sheets for twice, also reflected by the gold mirror, the laser beam was finally collected by the detector 1 for measurement, as shown in FIGURE 2(a). By gradually increasing the pump power, the power transmittance as a function of pump intensity was recorded and shown in FIGURE 2(b). To obtain the nonlinear optical parameters, a simple saturable absorption model of $T(I) = 1 - \alpha_0 \times \exp(-I/I_{sat}) - \alpha_{ns}$ [32] was used to fit the measured data. $T(I)$ is the transmission, α_0 is the modulation depth, I is the input intensity, I_{sat} is the saturation intensity, and α_{ns} is the non-saturable absorbance. Consequently, the modulation depth α_0, non-saturable loss α_{ns}, and saturation intensity I_{sat} were calculated to be 2.5%, 16.6%, and, 0.82 MW/cm^2, respectively.

III. EXPERIMENTAL SETUP

A simple linear cavity is constructed for achieve high-energy mode-locked TDFL, as shown in FIGURE 3. The linear cavity comprises a piece of 12-cm long highly doped single-cladding silica-based Tm^{3+}-doped fiber (core/cladding diameter = 5/125 μm, NA = 0.24) with the maximum absorption of $\sim$ 350 dB/m at $\sim$ 1550 nm in core, a single-moded WDM coupler (1550/2000 nm), and a 5.2-m-long SMF-28 fiber. The dispersions of the SMF-28 fiber and the Tm^{3+}-doped fiber at 1.9 μm are -67 and -12 ps^2/km [37], respectively. The total fiber cavity length is 5.32 m and the accumulated net cavity dispersion is around -0.7 ps^2. The pump source is a continuous wave (CW) erbium/ytterbium-codoped fiber laser with the maximum output power of around 1 W at 1550 nm, and this pump light was injected into the cavity via a wavelength division multiplexer (WDM) with a coupling efficiency of >98%. Moreover, an extrusion-type polarization controller was used to adjust the state of polarization of the laser.

In FIGURE 3, a gold mirror (coated with the WS_2 nanosheets) was precisely placed perpendicular to the cleaved fiber facet to butt-couple the guiding light to interact with the WS_2 nanosheets. At the other side of the TDFL, a cleaved fiber end provides a $\sim$ 4% Fresnel reflection and it, together with the gold mirror, can thus form the laser resonator. The gold mirror coated with WS_2 used as the SA, was mounted on a 5-dimensional stage (3 dimensions for linear translation

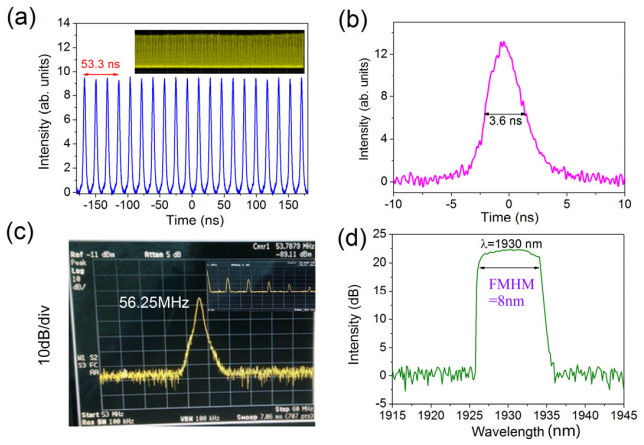

FIGURE 4. Characteristics of the 3rd order harmonic mode-locked TDFL using WS_2. (a) Pulse trains in the time domain. The inset picture shows the pulse trains over a wider time span of 5 μs. (b) Zoon-in time domain signal for a single pulse. (c) RF spectrum signals. The inset picture shows the whole RF spectrum spanning over 0-300 MHz. (d) Spectral characteristics of the mode-locked spectrum.

and 2 dimensions for rotation) for fine position adjustment. The laser pulses from the cavity were monitored using a high-speed InGaAs detector (Newport 818-BB-51, 12GHz Bandwidth), a 2.5 GHz sampling rate oscilloscope (Agilent, DSO9254A), and a radio frequency analyzer (Agilent, E4402B). The spectral responses were measured by a mid-infrared spectral analyzer (SIR 5000, SandHouse Co.) under a resolution of 0.22 nm and the laser output power was measured with a power meter (Thorlabs Co.).

IV. EXPERIMENTAL RESULTS AND DISCUSSIONS

The laser characteristics are shown in FIGURE 4. The mode-locked TDFL self-started with harmonic mode locking at a pump power of near 400 mW by finely adjusting the fiber end's butt position and direction with respect to the gold mirror. In contrast to conventional harmonic mode-locking where the harmonic number changes with pump power, the harmonic frequency of the WS_2 mode-locked fiber laser in this work remains the same (3rd harmonic) when the pump was increased to 950 mW from 400 mW or was decreased to 80 mW from 400 mW. With the pump power below 80 mW, the mode-locked lasing turns out to become a CW laser. The reason why the order of the harmonic mode-locked pulses remains the same with increasing pump power is ascribing to the high laser output coupling ratio (96%) and which results in a smaller cavity quality factor and a higher cavity loss in cavity [14].

The harmonic mode-locked pulses were readily obtained when the pump power was reached to 850 mW. The laser characteristics are shown in FIGURE 4 to monitor the stability. The pulsewidth was measured to be 17.8 ns which is corresponding to one third of the cavity round trip time (the total fiber length is ~10.6 m). It explicitly shows that the TDFL is operating at the 3rd harmonic mode-locking state and the inter-pulse intensity fluctuation is about 5%, as shown in FIGURE 4(a). Neither pulse breaking nor pulse-pair generation were observed. The characteristic for the single laser pulse under the maximum pump power of 950 mW is shown in FIGURE 4(b). The pulsewidth was measured to be around 3.6 ns with a near-Gaussian profile. The radio frequency (RF) spectrum was measured using an Agilent spectrum analyzer (E4402B) with a resolution of 1 kHz, as shown in FIGURE 4(c). The repetition rate for the mode-locked pulses is 56.25 MHz which is corresponding to the third harmonic mode-locking of a laser ~10.6-m-long cavity length. The signal-to-noise ratio was as high as 45 dB due to the clear mode locking situation. The corresponding harmonic mode-locked spectrum, shown in FIGURE 4(d), was recorded by a mid-infrared (mid-IR) spectrum analyzer (Avesta, Russia, ASP-IR-3.5) under a resolution of 0.22 nm. The Full Width at Half Maximum (FWHM) is 8 nm and the central wavelength λ is 1930 nm for the mode-locked spectrum. Based on the working parameters, the total net dispersion was calculated to be around -0.7 ps^2 and the mode-locked TDFL is supposed to be operating under the traditional soliton regime. However, as can be obviously found in FIGURE 4(d), the steep edges of the mode-locked spectrum clearly reflect that the TDFL is operating under the dissipative soliton regime. Though the group velocity dispersion for all the fibers is negative everywhere in cavity, the pulses were still highly chirped to result in a big time-bandwidth product of 2359. A similar result with a big time-bandwidth product was also proposed for a nanosecond mode-locked TDFL based on NPE effect [14]. The chirping is believed to be generated from the WS_2 since the absorption changes quickly with wavelengths over the 1.93 μm band, as shown in FIGURE 1(e). Based on Kramers-Kronig relations, a highly wavelength-dependent absorption loss will induce the high refractive index dispersion [41] to give rise to large chirping for the mode-locked pulses in this work. In addition, the strong absorption near the 1.93 μm wavelengths can also produce the issue of heat dissipation. Based on the experimental results, the environmental stability is a problem at this stage and the mode-locked pulses can be stable for less than 15 minutes contingent upon the concentration and thickness of WS_2 as well as the total cavity dispersion. Consequently, a temperature controlling device placed against the WS_2-coated mirror and a high precision actively-damped optical table could be highly helpful to substantially stabilize the WS_2-based mode-locked TDFL to last for a longer time in our future work.

FIGURE 5 shows the characteristics of the output power of the 3rd harmonic mode-locked 1.93 μm TDFL. The output power increases almost linearly with increasing pump power, and the maximum output power was 167 mW and the slope efficiency η is 61.9% with respect to the injected pump power. Because the frequency of the mode-locked TDFL was not changed, the pulse energy was increased with the increasing pump power. With the maximum pump power of 950 mW, the pulse energy was ~ 3 nJ. Clearly, the pulse energy was much higher than that of those lasers achieved using graphene or carbon nanotubes [28], [29]. Undoubtedly, the two-dimensional materials like WS_2 has great potentials

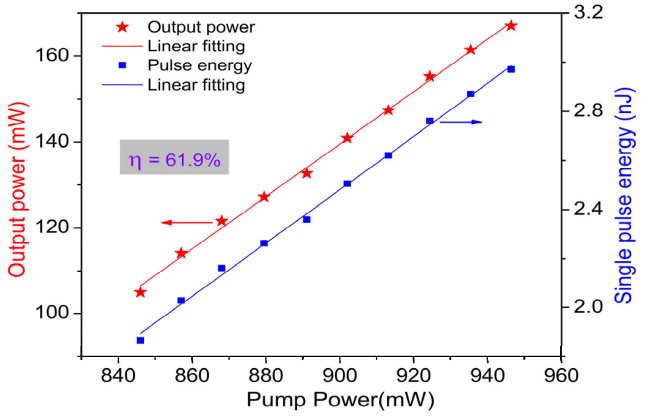

FIGURE 5. **Output power and pulse energy as a function of pump power.**

for the applications in mid-infrared high power and high energy lasers. Particularly, the 2 μm nanosecond pulsed lasers are essential for many important applications in micromachining, material processing, medical surgery, telehealth care, and environmental pollutant detections [42]–[44].

V. CONCLUSION

In conclusion, we have demonstrated the harmonic dissipative soliton mode-locked 1.93 μm TDFL based on multilayer WS_2 nanosheets using a compact linear cavity in anomalous dispersion regime. The harmonic mode-locking operation with the 56.3 MHz repetition rate can generate the pulse energy of up to 3 nJ. The pulsewidth is 3.6 ns while the FWHM of the mode-locked spectrum is 8 nm. An intriguing phenomenon was that the order of harmonic mode-locked pulses in the cavity remains the same (3^{rd} harmonic) when the pump was varying. In addition, the experiment results also reflect that the HDS can occur in the anomalous dispersion regime. The combination of high nonlinearity and large cavity dispersion can stabilize the frequency of harmonic mode-locking. Thus, the few-layer WS_2 is highly promising for serving as the saturable absorber for high frequency and high energy mode-locked fiber laser at the 2 μm wavelength range.

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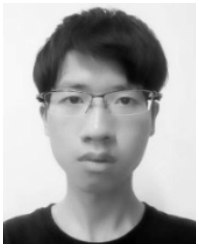

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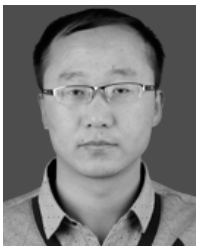

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