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Erbium-doped fiber laser passively mode locked with few-layer WSe₂/MoSe₂ nanosheets

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Few-layer transition-metal dichalcogenide WSe₂/MoSe₂ nanosheets are fabricated by a liquid exfoliation technique using sodium deoxycholate bile salt as surfactant, and their nonlinear optical properties are investigated based on a balanced twin-detector measurement scheme. It is demonstrated that both types of nanosheets exhibit nonlinear saturable absorption properties at the wavelength of 1.55 μm. By depositing the nanosheets on side polished fiber (SPF) or mixing the nanosheets with polyvinyl alcohol (PVA) solution, SPF-WSe₂ saturable absorber (SA), SPF-MoSe₂ SA, PVA-WSe₂ SA, and PVA-MoSe₂ SA are successfully fabricated and further tested in erbium-doped fiber lasers. The SPF-based SA is capable of operating at the high pump regime without damage, and a train of 3252.65 MHz harmonically mode-locked pulses are obtained based on the SPF-WSe₂ SA. Soliton mode locking operations are also achieved in the fiber laser separately with other three types of SAs, confirming that the WSe₂ and MoSe₂ nanosheets could act as cost-effective high-power SAs for ultrafast optics.

Two-dimensional layered materials have attracted increasing attention from fundamental researches to industrial applications attributing to their unique dimensionality effect as well as outstanding physical/chemical property^{1–4}. Graphene, the most famous two-dimensional material, possesses a Dirac-like electronic band structure, enabling ultra-broadband response ranging from ultraviolet to terahertz⁵. Since the few- and single-layer graphene was fabricated by the micromechanical cleavage method, it has been in-depth studied and applied in fields of photon detection, graphene plasmonics, ultrafast optics, quantum electrodynamics, *et al.*^{6–9}. However graphene also suffers the deficiency of the weak absorption coefficient (2.3% of incident light per layer), which limits its modulation ability and potential applications. In addition to graphene, transition-metal dichalcogenides (TMDs), a family of inorganic two-dimensional layered materials with the chemical formula of MX₂, where M is a transition metal (usually Mo, W) and X is a group VI element (S, Se, Te), are attracting continuously rising interest due to its exotic physical properties such as non-zero bandgap and layer-dependent second-order optical nonlinearity^{10–13}. The in-plane atoms of TMD materials are held together by strong chemical bonds while weak Van der Waals interaction enables stacking between layers, which enables them to be exfoliated into thin nanosheets for developing high-performance optoelectronic devices^{14,15}.

Molybdenum disulfide (MoS₂) and tungsten disulfide (WS₂), two representative TMDs, received the most research attention during the past several years^{16–18}. In the branch of optics, a great deal of research interest is focused on the nonlinear saturable absorption property of MoS₂ and WS₂ nanosheets^{19–23}. Wang *et al.* reported that few-layer MoS₂ display ultrafast saturable absorption property at 515 nm and 800 nm²⁰, and observed the strong two-photon absorption and saturable absorption respectively in mono- as well as multi-layer MoS₂ at 1030 nm²⁴. By the introduction of sulfur defects in MoS₂, Yu *et al.* found that the bandgap of MoS₂ nanosheets can be significantly reduced, and realized a broadband saturable absorber (SA) achieving Q-switched lasers at 1.06, 1.42, and 2.1 μm²¹. Recently, several groups including us demonstrated that WS₂ nanosheets exhibit broadband saturable absorption property even at sub-bandgap photon energies^{25,26}. Hereafter, various types of MoS₂ or WS₂ SAs were proposed to realize passively mode-locked or Q-switched lasers^{26–29}.

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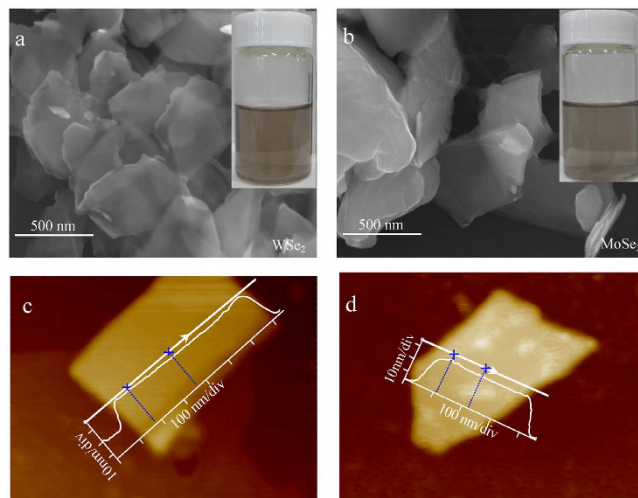


Figure 1. Characterization of WSe₂/MoSe₂ nanosheets. SEM images (a,b) and AFM images (c,d). The inset of Fig. 1(a,b) shows the corresponding suspensions of the nanosheets.

In comparison with MoS₂ and WS₂, molybdenum diselenide (MoSe₂) and tungsten diselenide (WSe₂) exhibit the similar molecular structure while have smaller optical bandgaps. For instance, the bandgaps of bulk MoS₂ and WS₂ are 1.29 and 1.35 eV while that of MoSe₂ and WSe₂ are 1.09 and 1.2 eV respectively³⁰. This special bandgap is particularly interesting for laser applications from the visible to near-infrared wavelength regime^{31–33}. Luo *et al.* demonstrated a compact red-light Q-switched praseodymium-doped all-fiber laser using two-dimensional TMDs as SAs³⁴. However, the study of WSe₂ and MoSe₂ is mainly concentrated on fabrication method and electronic property^{32,35} and their nonlinear optical behavior still remains less addressed up till now. We think that the investigation of emerging saturable absorption material is a key step for developing its potentials. In this contribution, we prepare WSe₂ and MoSe₂ nanosheets with a liquid exfoliation method using sodium deoxycholate bile salt as surfactant. Experimental results demonstrate that both WSe₂ and MoSe₂ nanosheets display saturable absorption property at the wavelength of 1.55 μm . By utilizing SPF- and PVA-based WSe₂/MoSe₂-SAs, stable picoseconds soliton pulses are delivered from the erbium-doped fiber (EDF) laser separately, including a train of 3252.65 MHz harmonically mode-locked ultrafast pulses.

Results

Preparation and characterization of WSe₂/MoSe₂ SAs. Various techniques have been explored to prepare mono- or few-layer TMD nanosheets, such as chemical vapor deposition, micromechanical exfoliation, liquid exfoliation, *et al.*¹¹. Each method has its advantages and ranges of applications. For example, micromechanical exfoliation is capable of producing high-quality nanosheets, while the production efficiency and size are limited. Chemical vapor deposition could synthesize large-scale mono- or few-layer TMD films. However, the synthetic process is quite complicated and requires high temperature. Liquid exfoliation is a simple and cost-effective method to produce mass dispersions of mono- and few-layer TMD nanosheets or other two-dimensional materials at ambient conditions^{36,37}. Here, the WSe₂/MoSe₂ nanosheets are prepared by the liquid exfoliation method, which is similar to that of previous reports³⁶.

The insets of Fig. 1(a,b) show the suspensions of WSe₂/MoSe₂ nanosheets, which display faint red color for WSe₂ and slight grey for MoSe₂ respectively. These suspensions are quite stable, and display no aggregation after being stored for several weeks under ambient conditions. The nanosheets are characterized by scanning electron microscope (SEM), as illustrated in Fig. 1(a,b). Both WSe₂ and MoSe₂ are present as two-dimensional nanosheets with intact surface texture, confirming successful exfoliation of the bulk-state crystal. The length and width of most nanosheets are below 1 μm , which depends on the centrifugation rate during the fabrication. The atomic force microscope (AFM) image illustrate that the thickness of nanosheets is ~ 20 nm for WSe₂ and ~ 15 nm for MoSe₂, as illustrated in Fig. 1(c,d). Based on above results, one can convince that the WSe₂/MoSe₂ nanosheets are successfully fabricated using the proposed method.

After the successful exfoliation of the bulk crystal, we then fabricate two types of WSe₂/MoSe₂-based SAs and investigate their saturable absorption properties. The first type of SA is obtained by depositing the WSe₂/MoSe₂ nanosheets onto a side polished fiber (SPF) to realize mutual interaction of nanosheets with the evanescent field of light within the SPF. The SPF is fabricated by polishing a bent single-mode fiber (SMF) on one side. The depth between the polished surface and fiber core of SPF is about 2 μm , and the polished part of the fiber has a length of 1.2–1.5 mm. Then the WSe₂/MoSe₂ nanosheets dispersion is dropped on the surface of SPF that is fixed on a quartz plate. At last, a 10 mW 1.55 μm continuous wave is coupled into the SPF and the optical deposition process starts. The deposited length and depth depend on the deposition time and input power, and the insertion loss is monitored in real time with an optical power meter. Actually, the insertion loss of the SA should be moderate, for example, mode locking operation can be achieved when the insertion loss ranges from 20% to 80%. By virtue of lateral interaction scheme, the TMD nanosheets interact with light over long distance while experience low intensity, which allows the SA to work at the high-power regime. Figure 2(a,b) shows the SPF deposited with

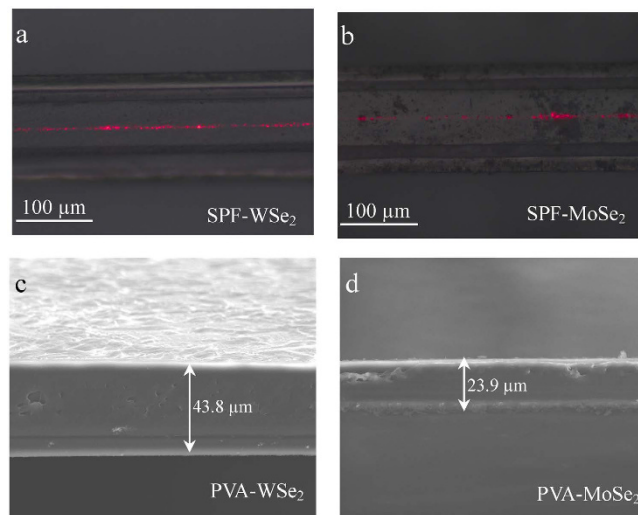


Figure 2. Optical micrograph of SPF deposited with WSe₂ nanosheets (a) and MoSe₂ nanosheets (b). Side profile of WSe₂-PVA film (c) and MoSe₂-PVA film (d).

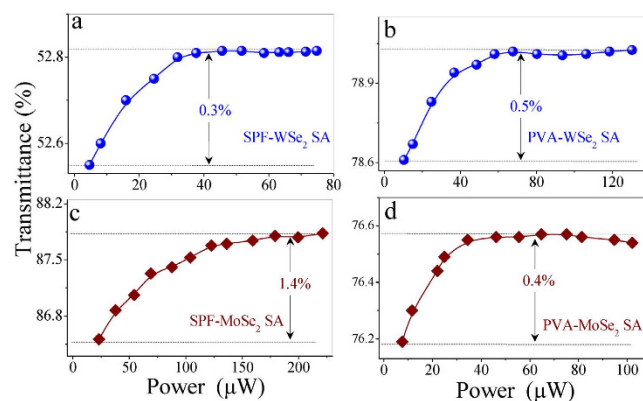


Figure 3. Nonlinear transmissions of SPF-WSe₂ SA (a), PVA-WSe₂ SA (b), SPF-MoSe₂ SA (c), and PVA-MoSe₂ SA (d).

WSe₂/MoSe₂ nanosheets after injecting a 632.8 nm He-Ne laser. The red laser is scattered from the surface of the SPF, indicating the existence of evanescent light field.

The other type of SA is fabricated by mixing the WSe₂/MoSe₂ nanosheets with aqueous PVA solution and evaporating the mixture on a substrate. This process could be described as follows. First, 5 Wt% PVA solution and the WSe₂/MoSe₂ dispersions are mixed at a volume ratio of 1:3 by a magnetic stirrer for 5 hours. Then, a thin WSe₂/MoSe₂-PVA film is formed by evaporating the mixture on a quartz plate under ambient temperature and pressure for 48 hours. At last, the film is cut into small pieces ($\sim 1 \times 1$ mm) and sandwiched between two facets of a fiber connector to realize fiber-based SA. The thicknesses of WSe₂/MoSe₂-PVA film are measured as 43.8 and 23.9 μm , as shown in Fig. 2(c,d), respectively. Compared with optical deposition³⁸ or dropping nanosheets on quartz plate³⁹, the PVA-film method would be more attractive due to its advantages of controllability, flexibility, and cost-effectiveness.

Based on a balanced twin-detector measurement method⁴⁰, we investigate the nonlinear saturable absorption property of four SAs (SPF-WSe₂, PVA-WSe₂, SPF-MoSe₂, and PVA-MoSe₂). Figure 3(a,d) shows the saturable absorption data of the WSe₂/MoSe₂ SA as a function of pulse power. One can observe that the transmission efficiencies of four SAs increase with pulse intensity, which is the typical characteristic of nonlinear saturable absorption. The modulation depths of SPF-WSe₂ and PVA-WSe₂ SAs are given as 0.3% and 0.5%, and that of the MoSe₂ are given as 1.4% and 0.4%. During the experiment, we did not observed nonlinear response from other devices, which suggest that the saturable absorption property is purely caused by TMD nanosheets. The proposed SAs may find wide applications in ultrafast photonics, including high-speed light modulation, optical switching, and ultrashort pulse generation.

Setup of WSe₂/MoSe₂ mode locked fiber laser. Fiber lasers possess inherent advantages of alignment-free operation, excellent beam quality, high efficiency^{41,42}, which provides an ideal platform to study the nonlinear optical property of new emerging materials such as carbon nanotube^{40,43}, graphene^{6,43,44}, topological

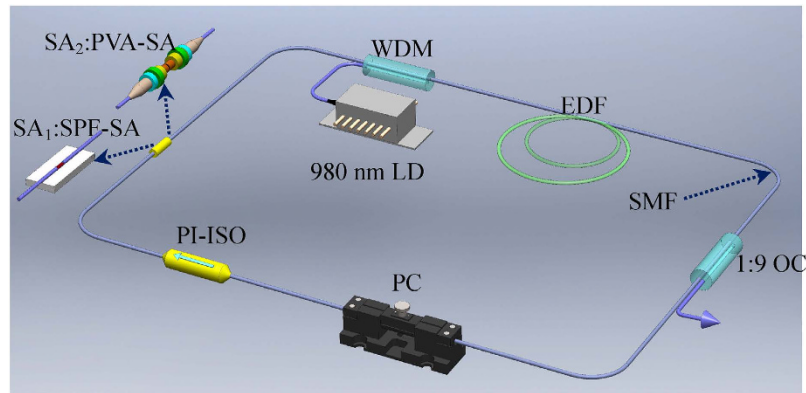


Figure 4. Schematic of the passively mode-locked fiber laser based on $\text{WSe}_2/\text{MoSe}_2$ SAs. SA_1 and SA_2 are SPF- and PVA-based SAs, respectively. Wavelength division multiplexer (WDM); Erbium-doped fiber (EDF); Output coupler (OC); Single-mode fiber (SMF); Polarization controller (PC); Polarization insensitive isolator (PI-ISO).

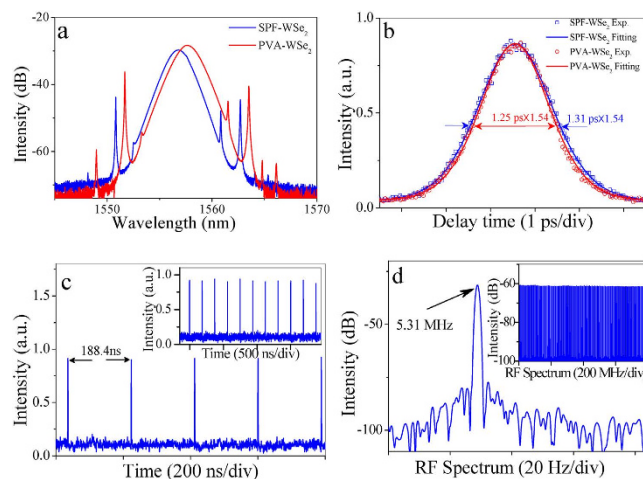


Figure 5. Output characteristics of WSe_2 mode-locked fiber laser. (a) optical spectrum, (b) autocorrelation trace, (c) pulse train, and (d) RF spectrum. The inset shows the broadband RF spectrum at a span of 800 MHz.

insulator³⁹, black phosphorus^{45,46}, and TMDs²⁸. Here, we construct an EDF laser to further test the saturable absorption property of four different SAs. The fiber resonator is composed of a 6 m EDF with 3 dB/m absorption at 980 nm, a fiber fused coupler with the output ratio of 10%, a polarization insensitive isolator, a SA, and a polarization controller, as shown in Fig. 4. The EDF used in the fiber laser is EDFC-980-HP, which has a mode field diameter of $5.8 \pm 0.5 \mu\text{m}$ and doping concentration of 1500 ppm. A 980 nm laser diode with the maximum power of 610 mW is used to pump the EDF via a wavelength-division multiplexer. The polarization controller can adjust the polarization state of the laser, but it is not fundamental to realize the mode locking operation. The pigtailed WSe_2 SA and MoSe_2 SA are 0.7 and 2.8 m, respectively. The other fiber is SMF with a total length of 32 m. Considering the dispersion parameters D of $-16 \text{ ps}/(\text{nm}\cdot\text{km})$ and $17 \text{ ps}/(\text{nm}\cdot\text{km})$ for EDF and SMF, the net dispersion β_2 of the fiber laser is calculated as -0.59 ps^2 for WSe_2 SA and -0.63 ps^2 for MoSe_2 SA, which facilitates soliton pulse shaping through the interaction of self-phase modulation and anomalous group velocity dispersion.

Experimental observations. We first investigate the mode-locking performance of the fiber laser by inserting the SPF- WSe_2 SA into the cavity. At the pump power of 21 mW, self-starting soliton mode locking operation is achieved with the output power of 0.45 mW. As illustrated by the blue curve in Fig. 5(a), the output spectrum is centered at 1556.7 nm with a 3-dB bandwidth of 2 nm. Several pairs of sidebands are symmetrically distributed in the optical spectrum, which is the typical characteristic of standard soliton. The pulse duration is measured by a high-sensitivity commercial autocorrelator. The autocorrelation trace has a full width at half maximum of 2 ps, as shown by the blue curve in Fig. 5(b). By using a Sech^2 fitting, the pulse duration is estimated to be 1.31 ps. During the experiment, the fiber laser tended to operate at multiple-pulses state due to the combined effects of soliton energy quantization and long cavity length⁴⁷. However, taking hysteresis phenomena of the soliton fiber laser, single-pulse mode locking can be obtained by slowly decreasing the pump power to 13 mW. The oscilloscope trace of the output pulse train is demonstrated in Fig. 5(c), which gives the pulse interval of 188.4 ns, well consistent with the cavity length of 38.7 m. The offset from zero position of y-axis on the oscilloscope trace may be

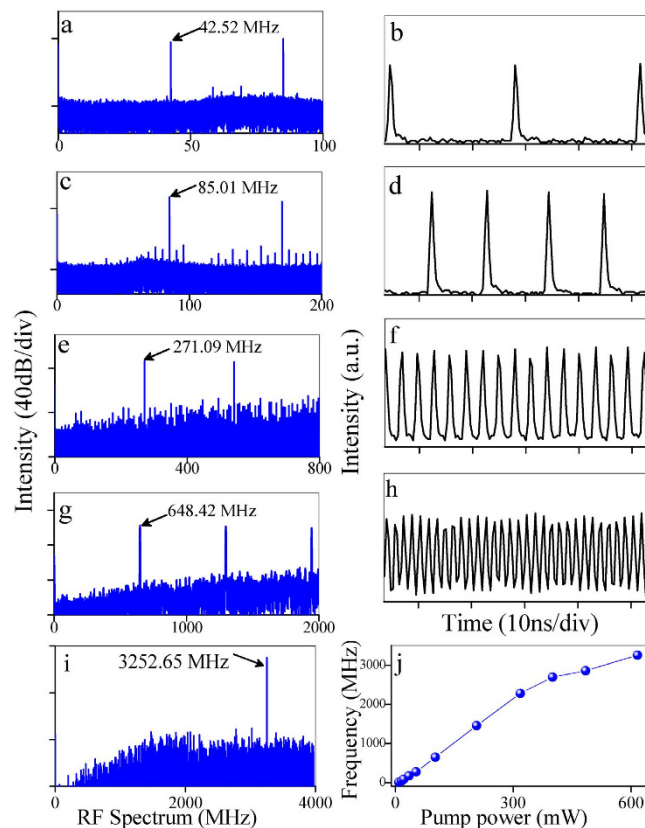


Figure 6. Harmonically mode-locked fiber laser based on the SPF-WSe₂ SA. RF spectrum (a,c,e,g,i) and pulse train (b,d,f,h); (j) Frequency of output pulses as a function of the pump power.

attributed to the combined effects of weak input power and continuous wave of the sidebands. The fundamental repetition rate of the pulse is given as 5.31 MHz, as shown in Fig. 5(d). The inset shows the broadband RF spectrum at a span of 800 MHz. To verify whether the mode locking is induced by WSe₂ nanosheets, we have replaced the SPF-WSe₂ SA by the PVA-WSe₂ SA in the cavity. In this case, soliton mode locking operation is also observed in the fiber laser at the pump of 35 mW. The optical spectrum and autocorrelation trace of the pulses are demonstrated by red curves in Fig. 5(a,b), respectively. Here, the output power of the fiber laser is measured as 0.84 mW, and the pulse evolution and radio frequency (RF) spectrum are quite similar with that of SPF-WSe₂ mode-locked fiber laser, which are not present in the text for conciseness.

Apart from the single- or multi-pulses operation reported previously, harmonic mode locking is also observed in the SPF-WSe₂ mode locked fiber laser. We use a 27.5 GHz RF analyzer with a 45 GHz photodetector to directly monitor the repetition rate of the output pulses. As shown in Fig. 6(a–i), by increasing the pump power from 25 to 610 mW, the repetition rate of the pulses gradually increases from 42.52 to 3252.65 MHz (612th harmonic of fundamental repetition frequency), and the output power increases from 0.6 to 19 mW. Due to the bandwidth limitation (500 MHz), the 3252.65 MHz pulses cannot be displayed correctly on the used oscilloscope. Figure 6(j) summarizes the evolution of the pulse repetition rate as a function of the pump power. The repetition rate almost changes linearly at low pump power. Actually, the maximum pulse repetition rate is limited by the available pump power and can be further enhanced by optimizing the SA, such as using fiber-taper based SAs. The formation mechanism of harmonic mode locking can be attributed to long-period interaction between solitons⁴⁸. When the solitons and non-soliton components exhibit certain phase difference, the interaction force becomes repulsive for all solitons within a pulse bunch and results in uniform distributed pulses inside the laser cavity. Such high-repetition-rate pulsed fiber laser can find important applications such as frequency combs and soliton communications.

As the extra heating can be rapidly dissipated from the SPF, the SPF-WSe₂ SA can work at mode locking state at the pump power of 600 mW during the whole experiment (about four hours). Using bidirectional pump scheme with the total pump power of 1.2 W, the fiber laser can work at mode locking state for several minutes and then evolves into continuous wave state. In this case, the intracavity power is calculated as 400 mW from the output power. However, by decreasing the pump power to zero and then increasing pump power, mode-locking operation can be obtained again. Based on these results, we infer that the damage threshold of the SA is about 400 mW.

By using SPF- and PVA-MoSe₂ SAs, mode-locked operation can also be realized in the fiber laser, similar to that of WSe₂ SA. The blue and red curves show the laser performances based on SPF- and PVA-MoSe₂ SAs respectively, as demonstrated in Fig. 7. The spectral width, pulse interval, pulse duration, and fundamental repetition rate for SPF-MoSe₂ mode-locked pulses are 2.3 nm, 1.09 ps, 198.8 ns, and 5.03 MHz, respectively. The

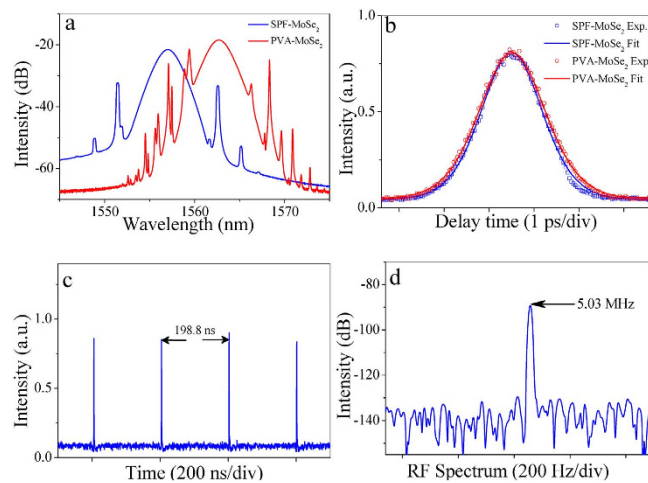


Figure 7. Output characteristics of MoSe₂ mode-locked fiber laser. (a) Optical spectrum, (b) autocorrelation trace, (c) pulse train, and (d) RF spectrum.

SA types	SA parameters		Nanosheets parameters		Mode-locked laser performance			
	Modulation depth (%)	Thickness (μm)	Size (μm)	Thickness (μm)	Central wavelength (nm)	Spectral Bandwidth (nm)	Pulse duration (ps)	Repetition Rate (MHz)
SPF-WSe ₂	0.3	//	0.4~1	~15	1556.7	2	1.31	5.31
PVA-WSe ₂	0.5	43.8			1557.6	2.1	1.25	5.31
SPF-MoSe ₂	1.4	//	0.4~1	~20	1557.1	2.3	1.09	5.03
PVA-MoSe ₂	0.4	24.9			1562.6	2.2	1.18	5.03

Table 1. Summarization of SA parameters and laser performance.

corresponding time bandwidth product is calculated as 0.316, indicating that the output pulse is a chirp-free soliton. The central wavelength changes for different mode-locking operations, which is mainly attributed to the variation of pump power, insertion loss, and polarization state. Table 1 summarizes the SA parameters and the corresponding laser performance for a clear comparison.

E. M. Pessina *et al.* demonstrated that self-mode locking operation can be achieved due to multimode laser instability⁴⁹. However, the instability-induced mode-locked fiber laser always operates at the fundamental repetition rate and cannot deliver stable picosecond pulses, which is different from our operations. For example, the MoSe₂/WSe₂ mode-locked fiber laser is capable of generating robust single/multiple pulses without average. Moreover, soliton mode locking operation could be also obtained in a fiber laser with length of 11 m using the proposed SAs. We have carried out comparative experiments to verify whether the mode locking operation is purely caused by WSe₂/MoSe₂ nanosheets. In the experiment, continuous wave is always observed in the fiber laser by using pure SPF (directly exposed on air), PVA film, or removing the WSe₂/MoSe₂ SAs, despite that the polarization controller and the pump power are tuned over a full range for hundreds of times. In contrast, mode-locking operation can be easily achieved by depositing the WSe₂/MoSe₂ nanosheets on SPF or inserting the PVA-WSe₂/MoSe₂ film into the laser cavity. The comparative results confirm again that the WSe₂/MoSe₂ SAs are responsible for the mode locking operation of the fiber laser.

Discussion

We found that the dispersion setting and output ratio of fiber cavity strongly affect the laser operations. For example, mode-locking operation can be easily obtained at large anomalous dispersion regime with output ratio less than 10%. However, pulse operation is difficult to achieve at normal dispersion regime or fiber laser with output ratio larger than 20%. The results could be attributed to the small modulation of the WSe₂/MoSe₂ SAs. As the normal dispersion induced temporal broadening or output induced-perturbation cannot be compensated by the SA, mode-locking will not be established in the fiber laser. Experimental experiments show that MoSe₂/WSe₂ nanosheets exhibit saturable absorption at 1.55 μm (0.8 eV), although the photon energy is lower than the bandgap of the material. Several groups have proposed different theories to interpret the sub-bandgap saturable absorption, such as defect-induced bandgap decreasing²¹, coexistence of semiconducting and metallic states²², two-photon absorption saturation²⁴, and edges state of the materials⁵⁰. However, a unanimity interpretation of the governing mechanism has not been fully established. In the view of defect-state theory, the deviation from perfection in two-dimensional materials is inevitable and will renovate their electronic and optical properties⁵¹. By the introduction of sulfur defects in MoS₂ or WS₂, the bandgaps of these materials can be significantly reduced^{21,25,52,53}, which may further contribute to the broadband saturable absorption. As these TMD materials possess

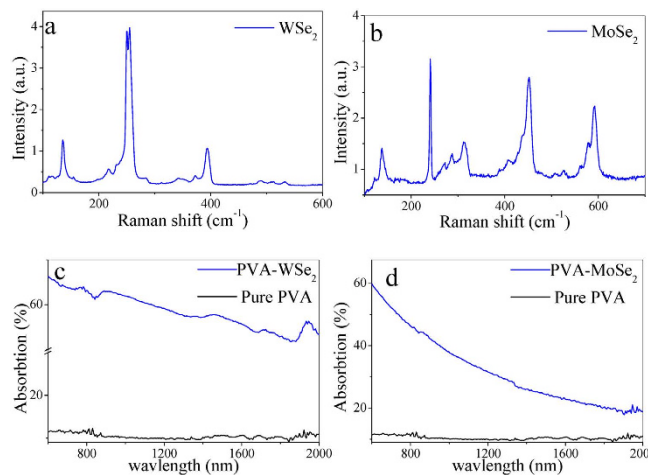


Figure 8. Raman spectra of (a) bulk WSe₂ and (b) bulk MoSe₂. (c) Linear absorptions of PVA-WSe₂ film and (d) PVA-MoSe₂ film in comparison with pure PVA-film.

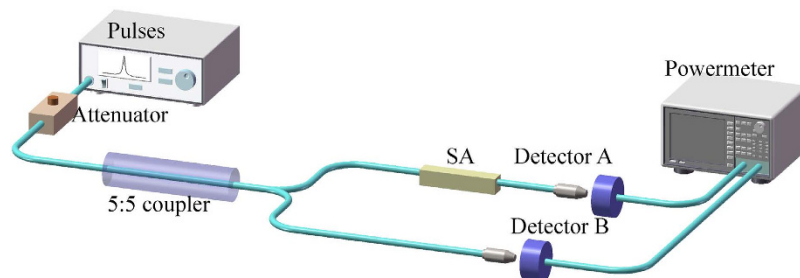


Figure 9. Schematic diagram of nonlinear optical characterization scheme.

similar lattice structures and photonic properties, we could deduce that sub-bandgap saturable absorption of WSe₂/MoSe₂ nanosheets may be similar with that of the MoS₂ and WS₂ reported previously.

Methods

Preparation of WSe₂/MoSe₂ nanosheets. Before the liquid exfoliation, we identify the WSe₂/MoSe₂ crystal by Raman spectroscopy using a 785 nm laser, as depicted in Fig. 8(a,b). The Raman spectra of bulk WSe₂ and bulk MoSe₂ agree well with the earlier findings⁵⁴. The WSe₂/MoSe₂ nanosheets are prepared by a liquid exfoliation method that consists of three steps. First, a solvent is prepared by mixing 50 mg sodium deoxycholate bile salt surfactant with 5 mL deionized water. Indeed, the solvent must be elaborately chosen to match the surface energy of the layered materials. Then, a piece bulk WSe₂/MoSe₂ crystal (about 15 mg) is dropped into the prepared solvent, and the mixture is ultrasonic treated for 8 hours at power of 180 W. Note that impurities and defects are inevitable in the prepared sample, which may induce a small bandgap and thus a broadband optical response²¹. To remove the large aggregation, the dispersion is further centrifuged at 1000 rpm for 10 minutes and the 90% supernatant is collected for the next step of the experiment. The linear absorptions of PVA-WSe₂ film, PVA-MoSe₂ film, and pure PVA film are measured by a spectrometer (Hitachi UV4100). As shown in Fig. 8(c,d), the absorption coefficient decreases with the increase of wavelength from the visible to near-infrared band.

Characterizing the nonlinear saturable absorption of WSe₂/MoSe₂ SA. The nonlinear saturable absorption property of SPF-WSe₂ SA, PVA-WSe₂ SA, SPF-MoSe₂ SA, and PVA-MoSe₂ SA are investigated separately with a balanced twin-detector measurement scheme. The illumination pulse is generated from a 1.55 μm EDF laser with the pulse repetition rate of ~25 MHz and the duration of ~400 fs. As shown in Fig. 9, the illumination pulse is divided equally by a coupler and the test sample is inserted in one branch. By adjusting the intensity of illumination pulse, the transmission of the SA as a function of incident power is obtained by comparing the output powers of two branches.

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Author Contributions

D.M. designed the work, performed the experiments, and wrote the paper. X.S. and B.D. tested saturable absorption of the devices. X.C. measured the property of WSe₂ and MoSe₂ nanosheets. W.Z. and K.S. performed the part of experiment. T.P. and B.J. carried out the data analysis and revised the manuscript. D.Y. and J.Z. contributed to the scientific discussion and considerably improved the manuscript presentation. All authors discussed the results and substantially contributed to the manuscript.

Additional Information

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