Research article

Lu Li*, Lihui Pang, Qiyi Zhao, Yao Wang and Wenjun Liu*

VS₂ as saturable absorber for Q-switched pulse generation

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Abstract: Transition metal dichalcogenides have been widely utilized as nonlinear optical materials for laser pulse generation applications. Herein, we study the nonlinear optical properties of a VS₂-based optical device and its application as a new saturable absorber (SA) for high-power pulse generation. Few-layer VS₂ nanosheets are deposited on the tapered region of a microfiber to form an SA device, which shows a modulation depth of 40.52%. After incorporating the microfiber-VS₂ SA into an Er-doped fiber laser cavity, passively Q-switched pulse trains could be obtained with repetition rates varying from 95 to 233 kHz. Under the pump power of 890 mW, the largest output power and shortest pulse duration are measured to be 43 mW and 854 ns, respectively. The high signal-to-noise ratio of 60 dB confirms the excellent stability of the Q-switching state. To the best of our knowledge, this is the first illustration of using VS₂ as an SA. Our experimental results demonstrate that VS₂ nanomaterials have a large potential for nonlinear optics applications.

Keywords: two-dimensional materials; saturable absorber; passively Q-switching; fiber laser.

1 Introduction

Nonlinear optical materials, as fundamental components of electronic and optoelectronic devices, play a key role in the field of advanced photonics [1–6]. In recent decades, widespread attention has been paid to investigate these promising materials, which has become a research hotspot [7–10]. Some low-dimensional nanomaterials with outstanding attributes of fast response, low cost, wide-band linear optical absorption, high optical nonlinearity and simplicity of integrating into an optical system have been proved to be effective nonlinear optical materials [11–16]. Particularly, two-dimensional (2D) materials are developed for wide applications in the nonlinear optical field because of their attractive photonic characteristics, including ultrafast carrier dynamics, strong light-matter interaction and large modulation depth. Consequently, the 2D materials provide a good platform for potential optical applications of, for example, optical modulation, optical limiting and photodetectors [17, 18].

As we all know, the pulse lasers have various applications such as basic physics, precision material processing and health care. Passively mode-locking and Q-switching techniques are the two main ways to generate pulses, whose prospective applications are on the rise [19–21]. Currently, graphene [22], topological insulators (TIs) [23], metal-organic frameworks [24, 25], perovskite [26], MXene [27], group-VA mono-elemental materials [28–30], transition metal dichalcogenides (TMDCs) [31, 32], black phosphorus [33–35] and IIIA/IVA monochalcogenides [36] have been extensively investigated and confirmed as great alternatives to saturable absorbers (SAs) in pulsed laser systems for ultrashort pulse generation. In the past few years, extensive efforts have been made to use TMDCs as SAs to induce Q-switching and mode locking over a wide wavelength range from visible to near infrared, which exhibits tunable bandgap, low optical attenuation and nonlinear optical effect generation [37, 38]. The general term TMDC is used to refer to the combination of chalcogen atoms (S, Se and Te) and transition metals (groups IVB to VIII, IB, and IIB). Actually, the group VIB TMDCs (MoS₂ and WS₂) have been used for pulse generation thoroughly

***Corresponding authors: Dr. Lu Li**, School of Science, Xi'an University of Posts and Telecommunications, Xi'an 710121, China, e-mail: liluyoudian@163.com. <https://orcid.org/0000-0002-3097-1954> and **Prof. Wenjun Liu**, State Key Laboratory of Information Photonics and Optical Communications, School of Science, Beijing University of Posts and Telecommunications, Beijing 100876, China, e-mail: jungliu@bupt.edu.cn. <https://orcid.org/0000-0001-9380-2990>

Lihui Pang: Shaanxi Provincial Center for Regenerative Medicine and Surgical Engineering, First Affiliated Hospital, Xi'an Jiaotong University, Xi'an 710061, China

Qiyi Zhao and Yao Wang: School of Science, Xi'an University of Posts and Telecommunications, Xi'an 710121, China

[39, 40]. Furthermore, some other TMDCs with a small bandgap, including ReS₂ and HfS₂, have also been used as SAs [41, 42]. However, the VB TMDC-based ultrafast photonics devices remain at the initial stage of development [43]. VS₂, as a representative conducting VB TMDC, consists of a metal vanadium sandwiched between the two sulfide layers as S-V-S, exhibiting metallic behavior with negligible bandgap [44]. Due to a wealth of intriguing properties of magnetism, charge density waves, and superconductivity, VS₂ has particular advantages for multifunctional applications in the field of supercapacitor, nanoelectronics, energy storage and hydrogen evolution [45, 46]. To the best of our knowledge, there has been no research devoted to the nonlinear optical property of VS₂.

In this study, we have prepared a microfiber-VS₂ device and used it as a new SA for high-power nanosecond Q-switched pulse generation. VS₂ nanosheet dispersion is fabricated via the liquid exfoliation method using N-methylpyrrolidone (NMP) as a solvent. Then, the VS₂ nanosheets are deposited on the tapered region of a microfiber to form an SA device, which shows the modulation depth of 40.52%. Based on VS₂ saturable absorption, passively Q-switched pulses are generated in the EDF laser. The maximum output power/minimum pulse duration is 43 mW/854 ns under the pump power of 890 mW. The signal-to-noise ratio (SNR) up to 60 dB confirms good stability of a Q-switched fiber laser. These experimental

results indicate that VS₂ can be used as a remarkable SA material for high-power pulse generation.

2 Experiments

2.1 Preparation of a microfiber-VS₂ SA

Top to down methods are the common techniques to make layered structure materials from bulk to few layers. Depending on the fabrication conditions, the nanometer sizes can be tunable. Figure 1A depicts the VS₂ lattice structure. The interval between two layers is 5.76 Å. Liquid phase exfoliation may be an ideal process to fabricate VS₂ nanosheets with the advantages of easy preparation and high quality. The detailed fabrication process was as follows. First, the bulk VS₂ crystal was ground into powder. The second step was the sonification procedure. The 10 mg VS₂ powder and 20 ml NMP solvent were mixed. Afterward, the mixed solution underwent ultrasonication for 8 h. The NMP has been demonstrated to be a good solvent for exfoliating 2D materials, whose surface energy matches well with VS₂ nanosheets [47]. So we do not utilize surfactant as the assisted medium. During this process, the VS₂ nanosheets with different thicknesses were chopped with the aid of sonication energy and

Figure 1: The characters of VS₂.

(A) The lattice structure of VS₂; (B) the photograph of VS₂ nanosheet dispersion; (C) Raman spectra of VS₂.

Figure 2: AFM image and the corresponding height profile.

temporarily stabilized by physically interacting with the NMP solvent. Few-layered VS_2 nanosheets were obtained via the next centrifugation process. The VS_2 nanosheet dispersion is shown in Figure 1B. Raman spectra are displayed in Figure 1C. The E_{1g} and A_{1g} active modes locate at 281 and 373 cm^{-1} , corresponding to in-plane and out-of-plane vibrations of VS_2 . Figure 2 shows the atomic force microscopy (AFM) image of VS_2 nanosheets. The cross-section height profile along four dotted lines indicates the thickness of VS_2 nanosheets ranging from 17 to 20 nm. The indirect evanescent field coupling approach is developed to construct the VS_2 -based SA device, which can realize stabilizing and strong light-matter interaction between light and VS_2 nanosheets. The microfiber- VS_2 SA is formed by depositing VS_2 nanosheets onto the side surface of a microfiber when the laser passes through the microfiber. The waist diameter of the taper is set to be $12\text{ }\mu\text{m}$ and the length of it is 2 mm. The taper waist of $12\text{ }\mu\text{m}$ leads to a strong evanescent-material interaction. The interaction length of 2 mm enables the VS_2 nanosheets take full advantages of the modulation ability.

2.2 Laser configuration

The laser structure is schematically shown in Figure 3. The pump source is a laser diode with output power adjusting

from 0 to 900 mW. A 980/1550 wavelength division multiplexer connects the pump source and resonant cavity. The length of the gain fiber (ER110-4/125, Thorlabs, Inc., Newton, NJ, USA) is selected to be 35 cm. The fiber polarization independent isolator is set to ensure the laser unidirectional transmission but not filter the polarization direction. The SA is a piece of microfiber coated with VS_2 nanosheets. The waist region is tightly straightened and fixed on the quartz flake. The output port is a 50/50 optical coupler and the pigtail fiber is about 50 cm SMF-28e fiber. The pigtail fiber of all optical elements is SMF-28e fiber and the total cavity length is about 3.9 m. The measuring instruments include the following: a 1 GHz bandwidth and 10GSa/s digital oscilloscope (Rohde & Schwarz RTO2014, Munich, Germany), a 13.6 GHz radio frequency (RF)

Figure 3: Schematic illustration of the Q-switched EDF laser based on the microfiber- VS_2 SA.

spectrum analyzer (Rohde & Schwarz FSW13, Munich, Germany), an optical spectrum analyzer (YOKOGAWA AQ6370D, Tokyo, Japan), and a fiber optical power meter (VIAVI OLP-85, JDSU, Breinigsville, PA, USA).

3 Results and discussion

3.1 The nonlinear optical absorption of SA

The linear transmittance of the microfiber-VS₂ SA is measured from 1500 to 1600 nm. As depicted in Figure 4A, the transmittance is 46.4% at 1550 nm. The balanced twin detector measurement is used to judge the nonlinear optical response characteristics of the microfiber-VS₂ device. A nonlinear polarization rotation mode-locked EDF laser acts as the optical source, which operates at 1556 nm with a repetition rate of 42 MHz and a pulse duration of 600 fs. Figure 4B shows the dependence of transmittance on the incident laser peak power density. According to the two-level saturable absorption model, the nonlinear optical absorption of the proposed SA device is fitted by the following equation:

$$\alpha(I) = \frac{\alpha_s}{1 + \frac{I}{I_{\text{sat}}}} + \alpha_{\text{ns}}$$

where the saturable intensity (I_{sat}), nonsaturable loss (α_{ns}) and modulation depth (ΔT) are calculated to be 18.06 MW/cm², 15.07%, and 40.52%, respectively.

3.2 Q-switched operation

In this experiment, Q-switching operation can be obtained under the pump power of 150–890 mW. Figure 5A shows

the Q-switched pulse train evolution depending on different pump powers. With increasing pump power, the pulse period and pulse width decrease gradually. Fixing the pump power at 890 mW, we measured the shortest pulse duration to be 854 ns, which is shown in Figure 5B. The related optical spectrum characteristics are depicted in Figure 5C. It can be seen that the spectral shape with the central wavelength of 1531 nm remains almost unchanged. While the 3-dB bandwidth widens to 4 nm slowly with the pump power increasing to 890 mW. Setting the resolution bandwidth to be 200 Hz, we measured the RF spectrum in the span of 510 kHz at the pump power of 890 mW. As demonstrated in Figure 5D, the repetition rate locates at 233 kHz with the SNR of 60 dB, confirming the good stability of this Q-switched fiber laser. Figure 5E depicts the typical features of the Q-switched fiber laser. The repetition rate is expanded from 95 to 233 kHz and the pulse width is lowered from 4.27 μ s to 854 ns as the pump power is increased from 150 to 890 mW. As shown in Figure 5F, in the pump range of 150–890 mW, the average output power increases from 3.58 to 43 mW almost linearly. The single pulse energy enlarges from 36.9 to 184.6 nJ monotonically. Under the pump power of 850 mW, we keep this Q-switched fiber laser working 2 h a day for 10 consecutive days. We note that the pulse trains remain stable with no appearance of pulse splitting, indicating that the long-term stability of working is good. It is widely accepted that the mode-locked pulse characteristics are determined by the interaction of dispersion and nonlinear effects. In this work, the dispersion and nonlinearity have not reached equilibrium. Besides, the 50/50 optical coupler is used. Therefore, the intra-cavity power is relatively low. Thus, the passive mode-locking phenomenon does not appear.

Table 1 demonstrates the output performances of passively Q-switched EDF lasers to compare the nonlinear optical response of VS₂-based SA with the same kind of SAs using other 2D materials. The modulation depth

Figure 4: The optical characteristics of microfiber-VS₂ SA.

(A) The linear transmittance of the microfiber-VS₂ SA; (B) the nonlinear optical absorption curve of the microfiber-VS₂ SA.

Figure 5: Experimental results. (A) The Q-switched pulse sequence at different pump powers; (B) single pulse duration at the pump power of 890 mW; (C) the optical spectrum; (D) the RF spectrum; (E) relationship of pump power and repetition rate (pulse width); (F) relationship of pump power and output power (pulse energy).

Table 1: Comparison of the performances of Q-switched EDF lasers based on different 2D SAs.

SA	ΔT (%)	Repetition rate, kHz	Pulse width, μs	Output power, mW	Pulse energy, nJ	Reference
Graphene	45	3.3–69.5	3.7	1.1	16.7	[48]
Black phosphorus	0.47	5.73–31.07	3.59	4.2	142.6	[49]
$\text{CH}_3\text{NH}_3\text{PbI}_3$	–	15.2–36.4	0.919	28	770	[50]
SnS	36.4	36.36–65.19	–	1.1	–	[51]
ReS_2	0.12	12.6–19	5.496	1.2	62.8	[52]
TiS_2	8.3	25.2–50.7	4	0.48	9.46	[53]
WS_2	2.9	90–125	–	5.7	46.3	[54]
TiSe_2	25.92	70–154	1.126	11.54	74.9	[55]
HfSe_2	6.65	12.97–45	4.5	7.5	167	[56]
MoSe_2	21.7	47.5–105.7	1.09	23.2	224	[57]
PtSe_2	4.9	20.5–79.2	0.92	11.34	143.2	[58]
VS_2	40.52	95–233	0.854	43	184.6	This work

of 40.52% is much larger than that of other SAs, which is originated from the evanescent field coupling mechanism. This scheme can increase the interaction length between light and VS₂ materials; therefore, the nonlinearity of the VS₂-based SA device would be enhanced. So the modulation of light is stronger. For the pulse duration comparison, the pulse duration of listed Q-switched fiber lasers is mostly in the μ s time scale, while ns-level Q-switched pulses are obtained in our experiment. The repetition rate range of 95–233 kHz and the maximum pulse energy of 184.6 nJ in the current work are competitive to that of previous reports. Notably, the maximum output power of 43 mW is much higher than that of passively Q-switched EDF lasers with other 2D materials. Overall, our results are superior to other works in terms of modulation depth, pulse duration, pulse energy and average output power. The saturable absorption process of VS₂ can be explained by the Pauli blocking principle. When weak-intensity incident light illuminates the VS₂ material, the light can excite electrons from the valence band into the conduction band. These photon-generated hot carriers cool down rapidly and form a hot Fermi-Dirac distribution. Thus the newly formed electron-hole pairs would suppress the inter-band photon transition. Next, the electron-hole pair recombination dominates until the distribution of electrons and holes returns to equilibrium. When the light intensity is strong enough, the photon-generated carriers rise immediately and occupy the energy states nearby the edge of the conduction and valence band completely. Because of the Pauli blocking principle, the photons pass through without loss.

4 Conclusion

In summary, we introduced the new 2D nanomaterial VS₂ as a SA into the EDF laser for the first time. VS₂ nanosheets coated onto the microfiber form the SA device simply with the modulation depth of 40.52%. The repetition rate can be adjusted in the range of 95–233 kHz. The SNR of 60 dB confirms the stability of the Q-switched fiber laser. The maximum output power and shortest pulse duration are 43 mW and 854 ns at the pumper power of 890 mW. This work illustrates that VS₂ is a promising candidate of SA for the pulsed fiber laser with excellent output performance.

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