




Optical properties and applications for MoS₂-Sb₂Te₃-MoS₂ heterostructure materials

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Two-dimensional (2D) materials with potential applications in photonic and optoelectronic devices have attracted increasing attention due to their unique structures and captivating properties. However, generation of stable high-energy ultrashort pulses requires further boosting of these materials' optical properties, such as higher damage threshold and larger modulation depth. Here we investigate a new type of heterostructure material with uniformity by employing the magnetron sputtering technique. Heterostructure materials are synthesized with van der Waals heterostructures consisting of MoS₂ and Sb₂Te₃. The bandgap, carrier mobility, and carrier concentration of the MoS₂-Sb₂Te₃-MoS₂ heterostructure materials are calculated theoretically. By using these materials as saturable absorbers (SAs), applications in fiber lasers with Q-switching and mode-locking states are demonstrated experimentally. The modulation depth and damage threshold of SAs are measured to be 64.17% and 14.13 J/cm², respectively. Both theoretical and experimental results indicate that MoS₂-Sb₂Te₃-MoS₂ heterostructure materials have large modulation depth, and can resist high power during the generation of ultrashort pulses. The MoS₂-Sb₂Te₃-MoS₂ heterostructure materials have the advantages of low cost, high reliability, and suitability for mass production, and provide a promising solution for the development of 2D-material-based devices with desirable electronic and optoelectronic properties. © 2018 Chinese Laser Press

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1. INTRODUCTION

Two-dimensional (2D) materials have exhibited great promise in microelectronics and optoelectronics since the discovery of graphene [1–9]. They possess special physical features, and can be used in the construction of various functional devices. Considered as one of the keys to modern technology, low-dimensional materials have been consistently reported and deeply investigated [10–20]. For example, carbon nanotubes (CNTs) and some 2D materials have been applied to the preparation of saturable absorption devices, photodetectors, and optical modulators [21–28].

As a saturable absorption material, CNTs are not sensitive to polarization, and have relatively high damage threshold and environmental stability. However, their scattering loss needs to be further reduced [29–31]. Graphene, which is cheap

and convenient to prepare, also has a high damage threshold and ultra-fast recovery time [2–4]. As a special zero-gap semiconductor, graphene nearly absorbs the light for each band, which makes it suitable as an ultra-wide-band saturable absorber (SA). However, the modulation depth and non-saturable loss should be optimized [32–35]. Topological insulators (TIs), the generic term for materials with topological electronic properties, have the advantages of excellent saturable absorption property, large optical modulation depth, and high third-order nonlinear refractive index. Their narrow bandgap enhances their ability to absorb the broadband spectrum, which has potential applications in ultrafast optics [36–38]. Nevertheless, their thermal damage threshold should be improved [36–38]. Transition metal dichalcogenides (TMDs) have been widely investigated in nonlinear optics after graphene [39–45]. To date, molybdenum disulfide (MoS₂), tungsten

disulphide (WS_2), and tungsten diselenide (WSe_2) have been proved to have ultrafast electron relaxation ability. TMDs can act as saturable absorption materials. They have higher damage threshold, but their nonlinear absorption properties remain to be enhanced [39–45]. Black phosphorus has direct bandgap, and the carrier mobility is high. Although its easy oxidation brings about a bit of difficulty for its direct application [46–49], there are plenty of methods to encapsulate and enhance its stability that have achieved satisfactory results [50,51]. In addition, some novel 2D materials have garnered substantial interest. For instance, few-layer antimonene and few-layer bismuthene have successfully realized mode-locking operation [52,53]; they inject new vitality into the study of ultrafast lasers.

In fact, saturable absorption materials are composed of a monolayer or multilayer of atoms arrayed in 2D space or quasi-2D space with a certain thickness. They can be modularized, and then stacked together without lattice mismatch in traditional heterostructures [54–56]. Because different 2D materials have different bandgaps and energy band structures, they can be customized with different band structures according to various requirements [57–62]. Here, we provide MoS_2 - Sb_2Te_3 - MoS_2 heterostructure materials to produce SAs with higher damage threshold and larger modulation depth. The optical absorption intensity and modulation depth of the MoS_2 - Sb_2Te_3 - MoS_2 heterostructures can be greatly increased due to the greater modulation depth and stronger optical nonlinearity of Sb_2Te_3 . Compared with that of graphene, the damage threshold of MoS_2 is higher, and the modulation depth of single-layer MoS_2 is greater than that of graphene. Thus, MoS_2 will not introduce extra non-saturation losses on account of the increase in modulation depth. The damage threshold and modulation depth of SAs are measured to be 14.13 J/cm^2 and 64.17%, respectively, in experiment. The fabricated heterostructure SAs are integrated into an erbium-doped fiber (EDF) laser to demonstrate the related optical properties through Q -switching and mode-locking operation. Both the theoretical and experimental results indicate that the MoS_2 - Sb_2Te_3 - MoS_2 heterostructure materials have excellent optical properties in photonic device applications.

2. RESULTS AND DISCUSSION

A. Heterostructure Material Characterizations

The magnetron sputtering technique is employed to fabricate the MoS_2 - Sb_2Te_3 - MoS_2 heterostructure SAs. The heterostructure materials were divided into three layers, and were composed of MoS_2 and Sb_2Te_3 . The structures of the fabricated SAs are presented in Figs. 1(a) and 1(b). The gold film, which serves as a highly reflective mirror, was grown on polished fused silica substrate 1 inch in diameter. The MoS_2 target (diameter 60 mm, thickness 5 mm, purity 99.99%), Sb_2Te_3 target (diameter 49 mm, thickness 3.5 mm, purity 99.99%), and gold target (diameter 50 mm, thickness 2.5 mm, purity 99.9%) were placed in the target chambers, and the pressure in the vacuum chamber was set to be 10^{-4} Pa during the process of film deposition. At first, the Au target was excited under direct current (DC) voltage for 5 min with the gold film thickness of 117 nm. Then the sputtered sulfur and molybdenum

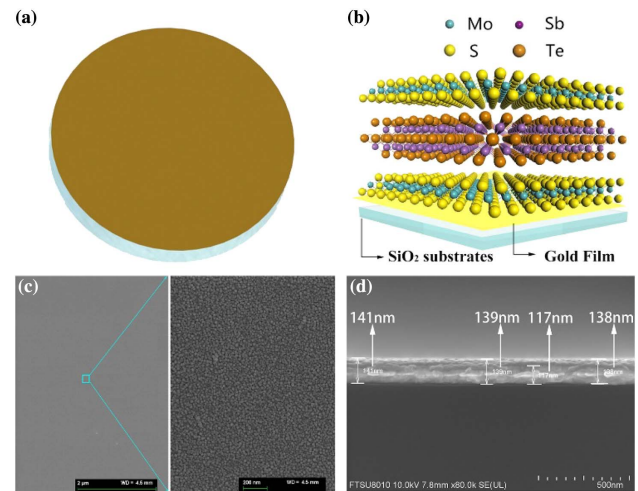


Fig. 1. State-of-the-art SA devices using the MoS_2 - Sb_2Te_3 - MoS_2 heterostructure. (a) Schematic of macrostructure and (b) surface structure of the fabricated MoS_2 - Sb_2Te_3 - MoS_2 heterostructure SA. Sb_2Te_3 (7 nm thickness) is in the middle of MoS_2 (8 nm thickness). The gold film with 117 nm thickness is deposited on the polished fused silica substrate as a broadband reflection mirror. (c) SEM image of the surface of deposited MoS_2 - Sb_2Te_3 - MoS_2 heterostructure film. (d) SEM image of the film thickness.

atoms were ejected out from the MoS_2 target and deposited on the gold film.

The MoS_2 film, with thickness of 8 nm, was generated under 100 W radio frequency (RF) power for 1 min. Next, Sb_2Te_3 film with thickness of 7 nm was deposited on the MoS_2 film under 0.2 A DC current for 20 s. Finally, another MoS_2 film with the thickness of 8 nm was deposited on the Sb_2Te_3 film. The size and microstructure of the MoS_2 - Sb_2Te_3 - MoS_2 heterostructure materials were verified by scanning electron microscopy (SEM). In Figs. 1(c) and 1(d), the surface of the heterostructure materials on the left side of the graph is compact and robust, and the as-prepared samples are composed of some uniform-scale nanoparticles on the right side of the graph, which demonstrate 8 nm average size. It is generally agreed upon that small particle diameter leads to large active surface area. Therefore, it can be speculated that MoS_2 - Sb_2Te_3 - MoS_2 heterostructures have outstanding activity and superior conductivity. The overall thickness of the heterostructure in Fig. 1(d) is about 24 nm.

B. Bandgap

The heterostructure was modeled by a MoS_2 - Sb_2Te_3 - MoS_2 sandwich-layered structure in Fig. 2(a). To minimize the lattice mismatch between the MoS_2 and Sb_2Te_3 , a supercell of $(\sqrt{7} \times \sqrt{7})/(2 \times 2)$ was used in Fig. 2(b), and the corresponding lattice mismatch is 2%. The corresponding unfolding band structure of the heterostructure was further examined with BandUP code [63,64]. The calculations were performed by density functional theory (DFT) implemented in the Vienna *ab initio* simulation package [65]. The interaction between core electrons and valence electrons was described with the projector augmented wave pseudopotential [66,67]. The valence electrons were expanded by plane wave functions with a 400 eV cutoff energy. The exchange-correlation energy was calculated by the use of the

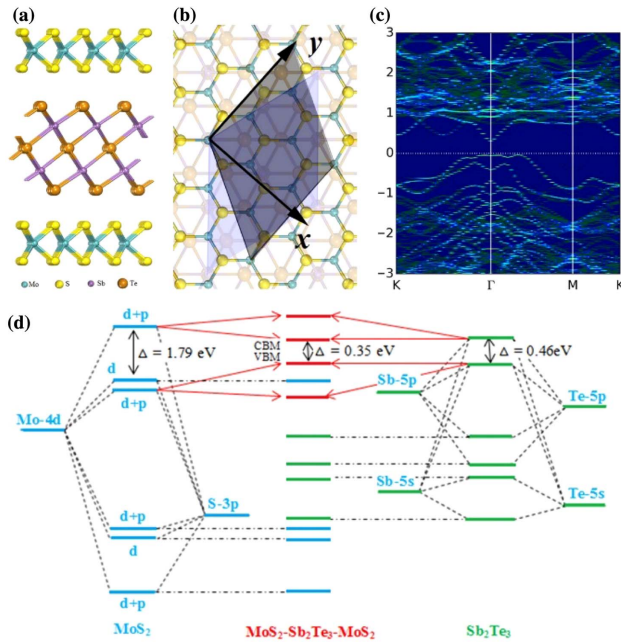


Fig. 2. Atomic and electronic structures of the MoS₂-Sb₂Te₃-MoS₂ heterostructure. (a) Side and (b) top views of the MoS₂-Sb₂Te₃-MoS₂ heterostructure. In (b), the detailed matching pattern of the $(\sqrt{7} \times \sqrt{7})/(2 \times 2)$ MoS₂-Sb₂Te₃-MoS₂ heterostructure is shown. The $(\sqrt{7} \times \sqrt{7})$ MoS₂ supercell is highlighted with yellow color, and the (2×2) Sb₂Te₃ supercell is denoted by the blue area. (c) Unfolding band structure of the MoS₂-Sb₂Te₃-MoS₂ heterostructure. Here, the Fermi level is defined as zero. (d) Band alignment of the MoS₂-Sb₂Te₃-MoS₂ heterostructure. The corresponding energy levels of pure MoS₂ and Sb₂Te₃ slabs are shown in both sides.

general gradient approximation of Perdew–Burke–Ernzerhof (PBE) [68]. Atomic spin-orbit coupling (SOC) was considered. The surface structures were modeled with a 20 Å vacuum separation between repeated slabs. To obtain the correct interlayer interaction in heterostructures, the van der Waals correction proposed by Grimme (DFT-D3) was enclosed [69]. By a mesh grid of $2 \times 2 \times 1$ k-points, the Brillouin zone was selected with the Gamma-centered Monkhorst–Pack scheme. The structure was relaxed until the change of the total energy was less than 10^{-5} eV and the forces became less than 0.01 eV/Å.

Bulk Sb₂Te₃ is a type of TI with a single Dirac cone. The calculated lattice constant of Sb₂Te₃ is 4.273 Å for a, and 29.975 Å for c using PBE + SOC. These calculated constants agree well with the experimental values: 4.264 Å for a, and 30.458 Å for c [70]. The lattice constant of MoS₂ is 3.150 Å for a, and 11.99 Å for c.

The modulation depth of the MoS₂-Sb₂Te₃-MoS₂ heterostructure is related to the carrier relaxation time τ and carrier concentration n_i . The relaxation time is associated with carrier mobility μ , and the μ in 2D materials can be calculated by the following equation [71,72]:

$$\mu_{2D} = \frac{e\langle\tau\rangle}{m} = \frac{e\hbar^3 C_{2D}}{k_B T m_e^* m_d (E_i)^2}, \quad (1)$$

where m_e^* (or m_h^*) is the effective mass along the transport direction (m_x along the x direction or m_y along the y direction,

respectively), which can be extracted from the band structure calculation, and m_d is the average effective mass defined by $m_d = \sqrt{(m_x m_y)}$. T represents the temperature taken as 300 K. E_i is the deformation potential constant of the valence band maximum (VBM) for the hole and conduction band minimum (CBM) for electron, defined by $E_i = \Delta E_i / (\Delta L_i / L_i)$, with the lattice compression or dilatation by a step of 0.5%. C_{2D} represents the effective 2D elastic moduli, which can be calculated by $C_{2D} = 2(E - E_0) / [S_0 (\Delta L_i / L_i)^2]$, where E and E_0 are the total energies after deformation along the i direction and at the equilibrium state, respectively. S_0 is the area of the 2D system at equilibrium. The intrinsic carrier concentration in 2D semiconductor materials can be computed through the following equation [73]:

$$n_i = \frac{\sqrt{m_h^* m_e^*} k_B T}{\pi \hbar^2} \exp\left(-\frac{E_g}{2k_B T}\right), \quad (2)$$

where E_g is the energy bandgap. In Fig. 2(c), the bandgap of the MoS₂-Sb₂Te₃-MoS₂ is calculated to be 0.35 eV, slightly smaller than the bandgap of monolayer Sb₂Te₃ (0.46 eV). As shown in Table 1, the effective masses of holes along the K - Γ (y) and Γ - M (x) directions in the heterostructure are $0.620m_0$ and $0.423m_0$, respectively. The corresponding effective masses of electrons are $0.315m_0$ and $0.405m_0$, respectively. The effective masses of holes are slightly larger than those of electrons along the same direction. Meanwhile, the effective mass of either electron or hole is smaller than that of the monolayer MoS₂, but a little larger than that of the monolayer Sb₂Te₃.

In Fig. 2(d), the bandgaps of the monolayers MoS₂ and Sb₂Te₃ are 1.79 and 0.46 eV, while that of the MoS₂-Sb₂Te₃-Sb₂Te₃ heterostructure is 0.35 eV. The main reason for this comes from the van der Waals interlayer coupling between antibonding $d + p$ hybrid orbitals of MoS₂ and $s + p$ hybrid orbitals of Sb₂Te₃, which causes the rise in VBM and the reduction of CBM. Meanwhile, such weak coupling also affects the carrier mobility and concentration, as we discuss subsequently.

C. Carrier Mobility

The effective mass and carrier mobility of the MoS₂-Sb₂Te₃-MoS₂ heterostructure and monolayers MoS₂

Table 1. Effective Mass (m_0) and Carrier Mobility (μ) of Monolayer and Heterostructure Materials^a

Units	Carrier Type	Effective Mass (m_0)		Carrier Mobility ($\text{cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$)	
		m_x^*	m_y^*	μ_x	μ_y
MoS ₂	e	0.774	0.478	23.87	94.04
	h	3.195	0.550	8.54	51.76
Sb ₂ Te ₃	e	0.291	0.157	0.75×10^4	1.91×10^4
	h	0.284	0.076	0.69×10^4	2.12×10^4
MoS ₂ -Sb ₂ Te ₃ -MoS ₂	e	0.405	0.315	1560.97	8474.63
	h	0.423	0.620	798.87	1447.54
Graphene-Sb ₂ Te ₃ -graphene	e	7.762	9.860	13.50	5.96
	h	1.553	1.656	149.23	357.62

^aHere, the monolayer MoS₂, monolayer Sb₂Te₃, MoS₂-Sb₂Te₃-MoS₂ heterostructure, and graphene-Sb₂Te₃-graphene heterostructure are considered.

and Sb_2Te_3 are shown in Table 1. The carrier mobility of the $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure is larger than that of the monolayer MoS_2 , but smaller than that of monolayer Sb_2Te_3 , which indicates that the relaxation time of $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ is longer than that of monolayer MoS_2 but shorter than that of monolayer Sb_2Te_3 . The carrier mobility of $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ is greatly affected by the relatively large effective mass compared with that of Sb_2Te_3 . In comparison with $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$, graphene- Sb_2Te_3 -graphene shows a larger effective mass, and thus the graphene- Sb_2Te_3 -graphene should have a shorter relaxation time than the $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$. The relatively large carrier mobility of the $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure represents the long relaxation time of the structure, which is vital for applications in photonic devices.

D. Carrier Concentration

The intrinsic carrier concentration, which is closely related with the effective mass and bandgap, is further calculated. As shown in Table 2, the carrier concentration of the $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ is 10–100 times larger than that of monolayer Sb_2Te_3 . The large carrier concentration of the $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure mainly comes from the small bandgap of 0.35 eV compared with that of monolayer Sb_2Te_3 . The large carrier concentration of the heterostructure indicates the small modulation depth. However, due to the small bandgap of 0.07 eV, the carrier concentration of the graphene- Sb_2Te_3 -graphene structure is about 10^3 times larger than that of the $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$.

E. Optical Property Characterization

Using the balanced twin-detector method as shown in Fig. 3, the modulation depth, saturation intensity, and nonsaturable absorption loss of the fabricated $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure SAs are measured. The pulse source is a homemade 125 MHz fiber laser centered at 1550 nm with 100 fs pulse duration and 80 mW output power. A variable optical attenuator (VOA) is used to control the power level of the optical pulses. A 50:50 optical coupler (OC) is applied to dividing the incident optical pulses into two paths with the same power. The main function of the circulator is to pass optical pulses through the SA mirror with a spatial structure. The SA mirror (SAM) is the fabricated $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure SA. By rotating the VOA, we can measure the different output powers from high to low at two detectors. Based on the measured data, we can fit the saturable absorption of the

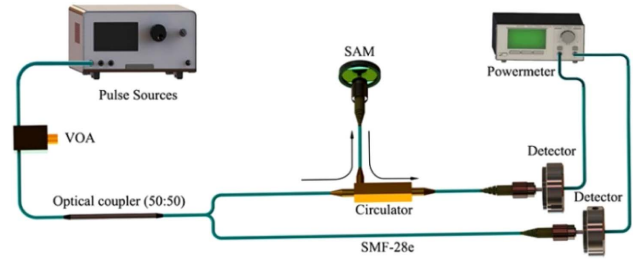


Fig. 3. Standard two-arm transmission setup. The SAM is the $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure SA mirror.

$\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure SAs, as described by the two-level model

$$\alpha(I) = \frac{\alpha_s}{1 + I/I_{\text{sat}}} + \alpha_{ns} \tag{3}$$

where α_s is the modulation depth (saturable loss), α_{ns} is the non-saturable loss, and I_{sat} is the saturation intensity. As presented in Fig. 4(a), the saturation intensity, non-saturable loss, and modulation depth are measured to be 151.176 MW/cm², 35.83%, and 64.17%, respectively. Such a large modulation depth considerably affects the performance of mode-locked fiber lasers. The large modulation depth of the $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure SAs is not only due to the larger modulation depth and stronger optical nonlinearity of Sb_2Te_3 in the heterostructure, but also related to the precisely controlled film quality in the manufacturing processes.

Using a Raman spectrometer which has the central wavelength of the laser around 514 nm, we measure the Raman spectra as shown in Fig. 4(b). In order to excite the Raman scattering, the laser power is set to be 5.6 mW. The peaks observed at 110, 118, 138, and 162 cm⁻¹ are well-related to the modes of Sb_2Te_3 . The peaks at 189 and 251 cm⁻¹ have relation with Sb_2O_3 . The peaks at 383 and 407 cm⁻¹ correspond to the E_{2g}^1 and A_{1g} modes of MoS_2 . Furthermore, owing to the direct interaction with high-power pulses, optically induced thermal

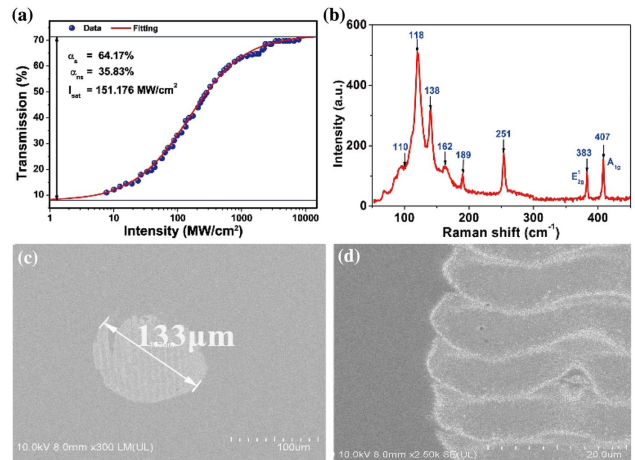


Fig. 4. Characterization of the $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure SA mirror. (a) The modulation depth is 64.17%. (b) Raman spectrum of the $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure. (c), (d) Threshold damage condition of the $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure film at 12 mW.

Table 2. Intrinsic Carrier Concentration of Monolayer and Heterostructure Materials^a

Units	Bandgap (eV)	Carrier Concentration (m ⁻²)	
		n_{ix}	n_{iy}
MoS_2	1.79	131	43.8
Sb_2Te_3	0.46	3.11×10^{12}	1.58×10^{12}
$\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$	0.35	5.13×10^{13}	1.03×10^{14}
Graphene- Sb_2Te_3 -graphene	0.07	7.26×10^{16}	1.13×10^{17}

^aHere, the monolayer MoS_2 , monolayer Sb_2Te_3 , $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure, and graphene- Sb_2Te_3 -graphene heterostructure are considered.

damage occurs in Figs. 4(c) and 4(d). Because of the thermal damage, long-term instability and pulse breaking arise, and the high-power operation of the fiber laser is limited. Thus, a high damage threshold for the SA is necessary. Here, the central wavelength of the light source for testing is mode-locked at 800 nm. The pulse duration and repetition rate are 35 fs and 1 kHz, respectively. The optical damage of the SA appears when the measured power is adjusted to 12 mW, corresponding to a 14.13 J/cm^2 damage threshold, which is four orders of magnitude higher than that of commercially available semiconductor saturable absorber mirrors ($500 \mu\text{J/cm}^2$) [39].

3. PULSED LASER APPLICATIONS

The schematic of the all-fiber EDF laser can be seen in Fig. 5. The length of the laser cavity is 5.6 m. The blue curve of the ring cavity represents the transmission fiber (SMF-28), and the green curve is the EDF as gain medium (Liekki 110-4/125). The length of the SMF-28 fiber is about 5 m, and the length of the high-gain EDF is about 60 cm. A polarization controller (PC) is engaged to change the polarization states of light, and a polarization-independent isolator (PI-ISO) is applied to controlling the unidirectional transmission of light in the laser cavity. The saturable absorber mirror is the fabricated $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure SA. The EDF laser is pumped by the laser diode (LD) at 976 nm, and the maximum output power of the LD is 600 mW. The pulse is output by 10:90 OC.

A. Q-Switching

After inserting the $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure SAs into the EDF laser cavity, a stable state of Q-switching is obtained in Fig. 6(a), which generates a series of stable pulse trains at 600 mW pump power. The central wavelength is around 1530 nm, as shown in Fig. 6(b). The pulse duration of full width at half maximum is about $1.6 \mu\text{s}$ in Fig. 6(c). Next, we measure its corresponding RF spectrum in order to investigate the stability of the fiber laser. In Fig. 6(d), the RF spectrum of the output pulse is presented, and the electrical signal to noise ratio (SNR) is about 42 dB, indicating high pulse train stability. The pulse trains without the amplitude fluctuation in Fig. 6(a) are complementary for the stability of our fiber lasers. Moreover, other frequency components are not observed in the RF spectrum with wider span in the inset, which further confirms the high stability of the fiber laser.

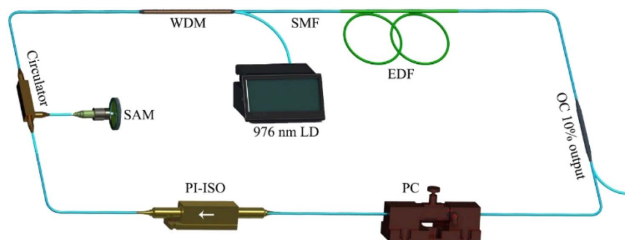


Fig. 5. Configuration of the mode-locked EDF laser. WDM, wavelength-division multiplexer; LD, laser diode; SMF, single-mode fiber; EDF, erbium-doped fiber; OC, optical coupler; PC, polarization controller; PI-ISO, polarization-independent isolator; SAM, $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure SA mirror.

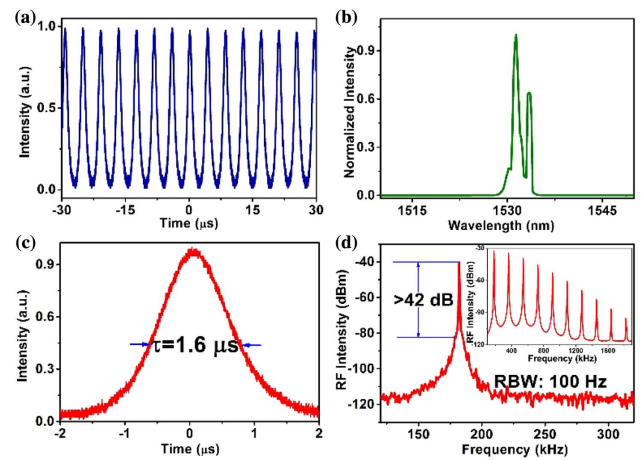


Fig. 6. Typical Q-switching characteristics. (a) Q-switched pulse trains. (b) Optical spectrum. (c) Q-switched pulse duration at 600 mW pump power. (d) RF spectrum at the fundamental frequency and wideband RF spectrum (inset).

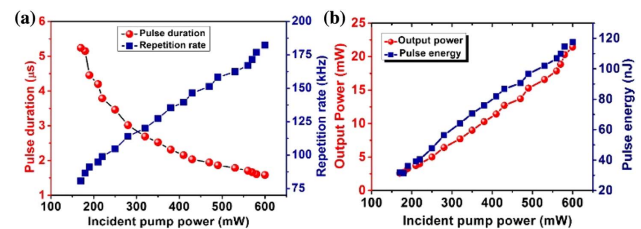


Fig. 7. (a) Pulse duration and repetition rate versus incident pump power. (b) Average output power and single pulse energy versus incident pump power.

The pulse properties of Q-switched fiber lasers mainly depend on nonlinear dynamics of the EDF, loss, and $\text{MoS}_2\text{-Sb}_2\text{Te}_3\text{-MoS}_2$ heterostructure SA. These lead to typical properties of the pulse duration and repetition rate versus incident pump power in Fig. 7. The pulse is emitted once the storage energy of the cavity reaches a certain threshold.

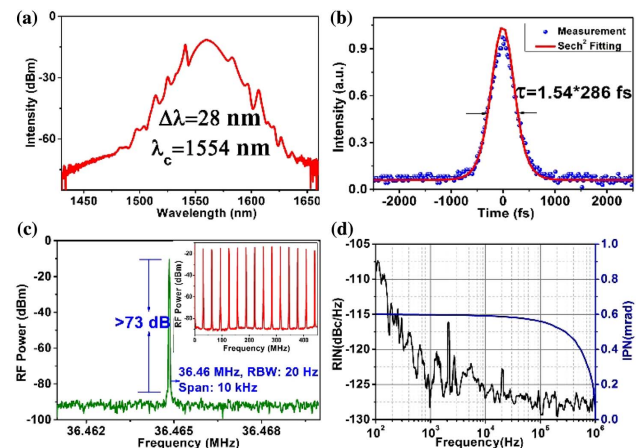


Fig. 8. Experimental results of fiber laser with mode-locked states. (a) Optical spectrum. (b) Pulse duration. (c) RF spectrum. (d) Phase noise.

Table 3. Comparison of Fiber Lasers Based on Different Heterostructure SAs

Materials	Pulse duration (fs)	SNR (dB)	Modulation depth (%)	Power (mW)	References
Graphene-Bi ₂ Te ₃ heterostructure	1800	67.4	18.98	–	[55]
Graphene-Bi ₂ Te ₃ heterostructure	837	60.7	12.6	3.07	[58]
Graphene-Bi ₂ Te ₃	189940	<50	23.28	2.53	[59]
MoS ₂ /graphene nanocomposites	3670	53.7	38.3	<2.16	[60]
Graphene/MoS ₂	830	60	10.8	5.85	[61]
Graphene/WS ₂	1120	62	9.6	4.74	[62]
MoS ₂ -Sb ₂ Te ₃ -MoS ₂	286	73	64.17	20	This work

When the incident pump power is adjusted from 180 to 600 mW, the pulse duration is reduced from 5.4 to 1.6 μ s, and the repetition rate increases from 87 to 182.3 kHz in Fig. 7(a). Therefore, higher repetition rates and shorter durations of pulses can be derived with the larger incident pump power. The relationship among the output power, pump power, and pulse energy is presented in Fig. 7(b). The maximum output power is 21.4 mW, and the highest pulse energy is 120 nJ. We believe that MoS₂-Sb₂Te₃-MoS₂ heterostructure SAs can endure higher power due to their homogeneous deposition. During the entire testing range of the pump power, the pulses with *Q*-switching states remain stable, suggesting the high quality and thermal stability of the heterostructure materials.

B. Mode Locking

Using a *Q*-switching system, the mode-locked laser pulses cannot be obtained by increasing the pump power and adjusting the PC in our experiment. We speculate that the nonlinearity and anomalous dispersion have not reached equilibrium in this case, so we are not able to get mode-locking operation. Taking into account the more negative dispersion meeting the increased likelihood of mode-locking, we have added a nearly 50 cm single-mode fiber between ISO and circulator on the basis of the *Q*-switching laser cavity. After fine-tuning the PC, we obtained mode-locked laser pulses. It is found that the stable mode-locking states appear at a relatively low mode-locking threshold value of 80 mW pump power. Once the mode-locking states of our fiber laser are realized, the fiber laser can remain stable when the pump power increases, without further adjustment of the PC. In Fig. 8, the related parameters are measured when the pump power is 600 mW. Figure 8(a) shows a typical soliton spectrum under the mode-locking state. The central wavelength is at 1554 nm, and the 3 dB bandwidth is about 28 nm. In Fig. 8(b), the pulse duration and maximum average output power are 286 fs and 20 mW, which are measured after the 50 cm single-mode fiber leading from the 10% output port of the OC. It is worth noting that the 286 fs pulse duration and 20 mW average output power reach the best level among the reported fiber lasers based on 2D material heterostructure SAs. Moreover, to the best of our knowledge, we demonstrate for the first time that MoS₂-Sb₂Te₃-MoS₂ heterostructures can be used as SA materials in fiber lasers.

To investigate the operation stability, we measure the RF spectrum. The repetition rate of the fiber laser is about 36.4 MHz. We perform 10 kHz frequency span spectrum to detect the fundamental repetition rate at the resolution bandwidth of 20 Hz in Fig. 8(c). There is no noise signal on the background, and the 73 dB SNR is observed, which

implies high stability in the mode-locking states. The strong inhibition ability for the noise may be related to the ultrafast electron relaxation of the MoS₂-Sb₂Te₃-MoS₂ heterostructure SAs. Besides, we show a wide RF spectrum span to observe the harmonic as a red line in Fig. 8(c). There is no significant spectral modulation, implying that the *Q*-switching instabilities do not exist. The integrated phase noise is measured in Fig. 8(d). The phase noise integrated from 100 Hz to 1 MHz is about 0.59 mrad, and the corresponding time jitter is about 2.9 ps. The related parameters of our fiber laser reach the best level among all fiber lasers based on heterostructure SAs (see Table 3).

4. CONCLUSION

MoS₂-Sb₂Te₃-MoS₂ heterostructure materials are prepared with magnetron sputtering technique, and successfully applied in ultrafast optics. The first-principles calculations reveal that the MoS₂-Sb₂Te₃-MoS₂ heterostructure can effectively improve the relaxation time and carrier concentration, compared with MoS₂ or Sb₂Te₃. The experimentally produced heterostructure SAs show 64.17% modulation depth and 14.13 J/cm² damage threshold. With the MoS₂-Sb₂Te₃-MoS₂ heterostructure SAs, *Q*-switching and mode-locking states are achieved in fiber lasers. With the incident pump power changing from 180 to 600 mW, we obtain tunable pulse duration from 5.4 to 1.6 μ s, and the repetition rate is changed from 87 to 182.3 kHz. For the *Q*-switched pulses, the maximum output power, SNR, and highest pulse energy are measured to be 21.4 mW, 42 dB, and 120 nJ, respectively. Due to the ultrafast electron relaxation of the prepared heterostructure materials, stable mode-locking pulses with 286 fs pulse duration and 20 mW average output power are also obtained. This work provides an alternative for preparing 2D materials with high quality and accurate controllability. Furthermore, the excellent optical properties of the prepared heterostructure materials indicate they can serve as promising materials for photonic devices.

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