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Molybdenum Disulphide Tape Saturable Absorber for Mode-Locked Double-Clad Ytterbium-Doped All-Fiber Laser Generation *

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We demonstrate the generation of passive mode-locked double-clad ytterbium-doped fiber laser operating in a 1-micron region. We prepare the saturable absorber from commercial crystal of molybdenum disulphide (MoS_2). Without chemical procedure, the MoS_2 is mechanically exfoliated by using a clear scotch tape. A few layers of MoS_2 flakes are obtained on the tape. Then, a piece of 1×1 mm tape containing MoS_2 thin flakes is inserted between two fiber ferrules and is integrated in the ring cavity. Stable mode-locking operation is attained at 1090 nm with a repetition rate of 13.2 MHz. Our mode-locked laser has a maximum output power of 20 mW with 1.48 nJ pulse energy. These results validate that the MoS_2 has a broad operating wavelength which covers the 1-micron region, and it is also able to work in a high-power cavity.

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In recent years, the high-power mode-locked fiber lasers that used two-dimensional (2D) layered material saturable absorbers (SAs) have attracted a considerable amount of attention. They offer many useful applications in various fields such as nuclear fusion, nonlinear optics, femtosecond laser nanomachining, medical, surgery, military and photonic sampling by utilizing high accuracy laser beams.[1,2] There are two techniques of making a mode-locked pulse, which are active and passive. The active mode-locking technique typically uses externally controlled devices such as acousto-optic modulators to actively modulate the intra-cavity light.[3] On the other hand, the passive mode-locking technique has not used an external signal. Instead, it is achieved by placing an SA or other passive element into the laser cavity which causes change in the intra-cavity light to generate pulsed laser.[3] Passive mode-locking can occur when an SA modulates the loss once per cavity round-trip dominant to the generation of a train of ultrashort (ps-fs duration) pulses at defined repetition rate corresponding to the free spectral range of cavity (MHz for few-meter fiber laser).[4] The passive mode-locking technique is compact, simple to construct and flexible in comparison with active mode-locking.[5] There are various types of SA such as nonlinear polarization rotation (NPR), semiconductor saturable absorber mirror (SESAM), carbon nanotube (CNT) and ion-doped crystal.[6] The NPR and SESAM are two commonly used SAs due to their high flexibility, stability, and fast amplitude modulation. However, SESAMs are more expensive, require complicated fabrications and packaging processes, and have a limited bandwidth of optical response.[7] Therefore, high-performance, low-cost and broadband operated SAs from other materials have gained attention of researchers in the field.

The most popular SA previously was graphene as it shows saturable absorption at very low optical intensities due to the 3rd order nonlinearity that exhibits wavelength-independent absorption.[6,8] Since then, mode-locked lasers have been demonstrated by using other 2D layered materials such as topological insulators (TIs),[9] black phosphorus (BP),[10,11] and transition metal dichalcogenides (TMDs).[12] They are considered as promising building blocks for the next-generation photonic technology due to their advantages such as having intrinsic property of broadband operation arising from its gapless linear dispersion of Dirac electrons.[6,13,14] TMD has attracted a considerable amount of attention due to its potential in many fields such as microelectronics, lithium batteries, medical and optoelectronics.[13,15] TMD is a group of 40+ layered materials with chemical structure of MX_2 with single plane of hexagonally-arranged transition metal (M) atoms covalently bonded together with two hexagonal planes of chalcogen (X) atoms.[4] TMD materials became popular mostly in the optical communication field due to their thickness-dependent band gaps,[16] their broadband saturable absorption,[17] high 3rd order nonlinearity susceptibility.[18] and ultrafast carrier dynamics.[19] The TMD mode-locked laser was reported first by Zhang *et al.*, in which the MoS_2 thin film was deposited on a fiber ferrule and integrated into a ytterbium fiber ring cavity.[17] These MoS_2 films are highly crystalline as their sizes can be up to several millimeters which can be used

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in electronics as transistors and in optics as emitters and detectors.[17] MoS_2 devices have been fabricated and characterized at 1 μm,[20] 1.5 μm,[2] and 2 μm,[21] which correspond to the gain bands. A strong SA has also been observed in other MoS_2-based devices such as MoS_2-coated fiber tips where modulation depths up to 32% and saturation intensities of 0.35 MW/cm^2 have been reported, although this was accompanied by very high non-saturable losses of ∼35.[2,4]

In this Letter, we demonstrate the fabrication of the MoS_2 2D-materials by the mechanical exfoliation method and its application as another stable SA for the high-power fiber laser using the double-clad ytterbium-doped fiber (YDF). The stable double-clad mode-locker is configured by sandwiching the MoS_2 film between two fiber connectors in the cavity. It exhibits stable mode-locked pulse and with the highest power obtained so far which makes MoS_2 a suitable candidate as an effective SA for the high power laser.

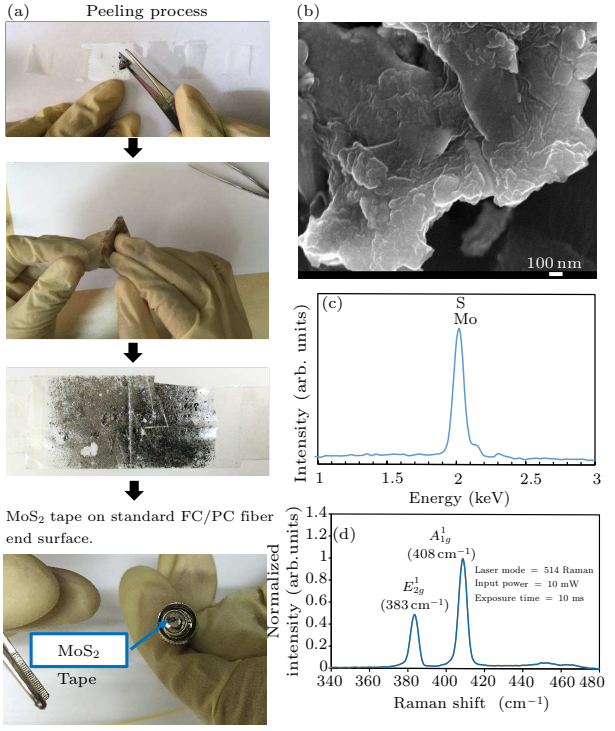

Fig. 1. (a) Mechanical exfoliation process. The obtained MoS_2 tape characteristics: (b) FESEM image, (c) EDX data, and (d) Raman spectrum.

In this experiment, the MoS_2 tape was prepared by the mechanical exfoliation method. This method has been widely used in graphene-based ultra-fast fiber laser application.[22,23] The advantages of mechanical exfoliation are due to its simplicity and reliability, where the entire fabrication process is free from complicated chemical procedures and costly instruments. Figure 1 shows the preparation process and material composition of MoS_2-tape-based SA. As shown in Fig. 1(a), relatively thin flakes are first peeled off from a large block of commercially available MoS_2 crystal (purity of 99%) by using a transparent scotch tape. Then the flakes are repeatedly pressed, thus MoS_2 flakes become thin enough to transmit light with high efficiency. The last step for preparation of MoS_2 is to cut a small piece of the MoS_2 tape, then attach it onto a standard FC/PC fiber ferrule end surface with index matching gel.

Figure 1(b) shows the field emission scanning electron microscopy (FESEM) image of the MoS_2 tape. The image confirms the absence of Mos_2 flakes or voids on the tape as small as 100 nm, which otherwise result in non-saturable scattering losses. It also indicates the existence of the uniform layers. The material composition of the MoS_2 tape was confirmed by the energy dispersive x-ray (EDX) analysis on the FESEM image, as shown in Fig. 1(c). At the energy of 2 keV, the Mo and S elements are dominant and verified to be in our SA tape. Raman spectroscopy was used to confirm the presence of MoS_2 and also estimates the number of layers.[24,25] Figure 1(d) shows the Raman spectrum, which was recorded by a spectrometer when a 514 nm beam of an argon laser with 10 mW exposure power was radiated onto the MoS_2 tape within 10 s. As shown in the figure, the sample exhibits two characteristic peaks, in parallel with two phonon modes; out-of-plane vibration of sulfide atoms at 408 cm^{-1}, and in-plane vibration of molybdenum and sulfide atoms at 383 cm^{-1}, with a frequency difference of 25 cm^{-1}. It is observed that the E^1_{2g} mode is present due to the in-plane motion red shifts after complete exfoliation, indicating that the few-layer MoS_2 with thicknesses of 5–6 layers had been successfully fabricated.[24,25] The full widths at half maximum (FWHM) of E^1_{2g} and A_{1g} bands are calculated to be 5.0 and 5.5 cm^{-1}, respectively, which agree with other reported results.[26]

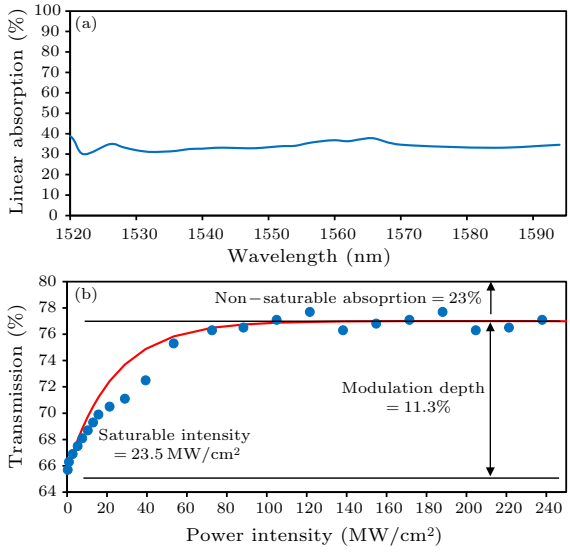

Fig. 2. Saturable absorber performances of the MoS_2 tape. (a) Linear absorption profile. (b) Modulation depth.

Another important parameter for SA is the linear

and nonlinear optical response. The amplified spontaneous emission (ASE) light source and mode-locked pulsed fiber laser are launched onto our MoS_2 tape to measure a linear and nonlinear optical response, respectively. Figure 2(a) shows the linear absorption of the MoS_2 tape within the range of 1520–1600 nm. The linear absorption is at the level of 35% and is characterized by a flat profile.

The nonlinear optical response property for the MoS_2 tape was investigated to confirm its saturable absorption via a transmission technique. Through balanced twin-detector measurement, the transmitted power is recorded as a function of the incident intensity on the device by varying the input laser power. The experimental data for transmission are fitted according to a simple two-level SA model of $T(I) = 1 - \alpha_s \exp(I/I_{sat}) - \alpha_{ns}$, where $T(I)$ is the transmission, α_s is the modulation depth, I is the input intensity, I_{sat} is the saturation intensity, and α_{ns} is the non-saturable absorption. The nonlinear transmission analysis of the MoS_2 tape is shown in Fig. 2(b). As shown in the figure, the modulation depth, non-saturable intensity, and saturation intensity are obtained to be 11.3%, 23.0%, and 23.5 MW/cm^2, respectively. This large modulation depth of 11.3% is expected to be able to suppress a wave breaking effect in the mode-locked fiber laser, and thus improves the attainable pulse energy.

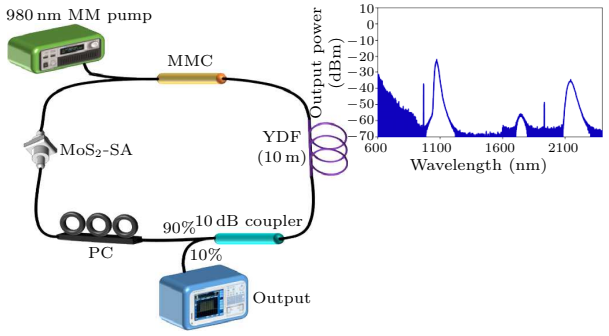

Fig. 3. Experimental setup of the mode-locked double-clad YDFL. The inset is the ASE image of YDFL.

A schematic diagram of the mode-locked double-clad ytterbium-doped fiber laser (YDFL) is illustrated in Fig. 3. The ring cavity consists of a 980 nm multimode pump, a multimode combiner (MMC), a 10-m-long double-clad ytterbium-doped fiber (Nufern LMAYDF10/130VII), a 10-dB coupler OC, and a polarization controller (PC). The YDF has a cladding absorption coefficient of 3.95 dBm at 975 nm and the group velocity dispersion (GVD) of -18 ps^2/km. Another fiber in the ring cavity is a standard SMF with a GVD of 44.2 ps^2/km. Thus the total net cavity dispersion is calculated under the normal dispersion condition to be $\sim$0.032 ps^2, which corresponds to the total cavity length of 14.8 m. The lasing is obtained after a multimode laser diode (LD) pump with emission centered at 980 nm was pumped into YDF via MMC. The YDF was employed as a gain medium, while MMC was used to combine pump and feedback signal together in a single optical fiber. Through the 10 dB coupler, 10% of lasing was coupled out as an output and 90% of lasing retained in the cavity. The polarization of oscillating light in the cavity was adjusted by using PC so that it matches the SA transmission axis. The size of 1×1 mm^2 MoS_2 tape was sandwiched between two fiber connectors' ferrule adhered with index matching gel. A stable pulse was obtained with some adjustment made manually at the fiber connectors that contain a piece of MoS_2 tape. Even though the cavity was pumped more than 1006 mW, this MoS_2 tape still worked effectively as SAs and is also proven to have high damage threshold characteristics. The lasing performance was observed by using an optical spectrum analyzer (OSA). Via the InGaAs high speed photodetector (PD), the 500 MHz oscilloscope and a 7.8 GHz rf spectrum analyzer were used to observe the temporal profiles of our mode-locked laser.

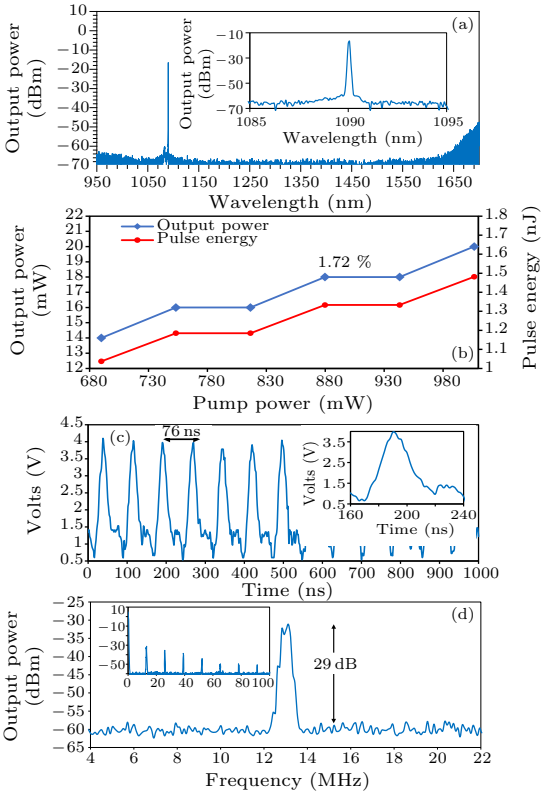

Fig. 4. Mode-locked laser performances: (a) output spectrum at the pump power of 689 mW, (b) output power and pulse energy, (c) pulse train at 1006 mW (inset: image of enlarge pulse train), (d) the rf spectrum at the maximum pump power of 1006 mW (inset: the rf spectrum with 100 MHz span).

The standard cw lasing of YDFL starts at 431 mW pump power. As we increase the pump power to 689 mW, the lasing starts to generate the mode-locked laser. The mode-locking operation remains stable un-

til 1006 mW and disappears as the pump power continues to increase. Stable mode-locking operation with a fundamental repetition rate of 13.2 MHz is generated between 689 mW and 1006 mW. Figure 4(a) shows the output spectrum of mode-locked YDFL which is obtained at the threshold pump power. The mode-locked output spectrum is induced at 1090 nm wavelength with 3 dB spectral bandwidth of 0.27 nm. Figure 4(b) shows the output power and pulse energy of the mode-locked YDFL with a slope efficiency of 1.72%. At a maximum pump power of 1006 mW, the measured output power is 20 mW and the maximum pulse energy is relatively calculated about 1.48 nJ.

Figure 4(c) shows the mode-locked YDFL pulse train with pulse period of 76 ns. The obtained fundamental repetition rate of 13.2 MHz matches the cavity length of 14.8 m. Directly obtained from the oscilloscope, the obtained pulse width of 32.5 ns is much broader than the actual pulse width. This happens due to resolution limitation of the oscilloscope. As an alternative, the pulse width can be measured by using an auto-correlator or calculation by using a formula based on the time bandwidth product (TBP). The minimum possible pulse width is estimated around 21.84 ps with TBP of 0.441 for Gaussian pulse profile and 3 dB spectral bandwidth of 0.08 nm. Figure 4(d) shows the rf spectrum with 100 MHz span. The repetition rate existence limits to seven harmonics and preliminarily indicates the pulse width is not too short. The rf spectrum confirms that the mode-locked YDFL has a fundamental repetition rate of 13.2 MHz with signal-to-noise (SNR) ratio of 29 dB. This SNR ratio is corresponding to the 10% amplitude fluctuation presence in the pulse train within one hour mode-locking operation. This small fluctuation is expected due to the inhomogeneous nature of MoS_2 flakes adhered on the scotch tape. The peak power is obtained to be about 0.154 kW. The long-term stability is also good since the pulse is stable for at least 48 h. The small changes in room temperature and environment do not affect the mode-locking performance of the laser.

In conclusion, we have experimentally demonstrated a passive mode-locking of 10 m double-clad YDFL with a MoS_2 tape as an SA. The stable mode-locked pulses are obtained from the threshold pump power of 689–1006 mW. The consistent pulse repetition rate of 13.2 MHz is obtained with SNR ratio 29 dB. At the pump power 1006 mW, the maximum pulse energy and output power are about 1.48 nJ and 20 mW, respectively.

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