# Passive harmonic mode-locking of Er-doped fiber laser using CVD-grown few-layer $MoS_2$ as a saturable absorber*

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Passive harmonic mode locking of an erbium-doped fiber laser based on few-layer molybdenum disulfide ($MoS_2$) saturable absorber (SA) is demonstrated. The few-layer $MoS_2$ is prepared by the chemical vapor deposition (CVD) method and then transferred onto the end face of a fiber connector to form a fiber-compatible $MoS_2$ SA. The 20th harmonic mode-locked pulses at 216-MHz repetition rate are stably generated with a pulse duration of 1.42 ps and side-mode suppression ratio (SMSR) of 36.1 dB. The results confirm that few-layer $MoS_2$ can serve as an effective SA for mode-locked fiber lasers.

**Keywords:** fiber lasers, mode-locking, molybdenum disulfide

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## 1. Introduction

Passively mode-locked fiber lasers are being used in a diverse range of practical applications including optical communication, laser micromachining, and optical frequency metrology.[1] To date, various techniques have been proposed to achieve passive mode-locking in fiber lasers, such as nonlinear-optical loop mirror (NOLM), nonlinear polarization rotation (NPR), semiconductor saturable absorption mirrors (SESAMs), and single wall carbon nanotubes (SWNTs). Since the first demonstration of graphene mode-locking,[2,3] graphene has been successfully exploited as an excellent saturable absorber (SA) for passive Q-switching and mode-locking applications in a broadband spectral range.[4–8] The success of graphene has greatly encouraged scientific researchers to explore other graphene-like two-dimensional (2D) nanomaterials, such as atomic layered transition-metal dichalcogenides (TMDs) for photonic applications.[9,10] 2D molybdenum disulfide ($MoS_2$) is a representative material of TMDs,[11] which is now attracting continuous attention due to its exceptional optical properties, including saturable absorption behavior[12] and ultrafast carrier dynamics.[13] Taking advantage of the excellent saturable absorption, 2D $MoS_2$ could be exploited as a promising SA for ultrafast laser applications. Recently, few-layer $MoS_2$ SAs have been successfully used in mode-locked fiber lasers. Zhang and colleagues presented $MoS_2$ mode-locked ytterbium-doped fiber lasers (YDFLs) producing dissipative solitons with pulse duration of 800 ps at 1.054 μm[14] and 656 ps at 1.042 μm,[15] respectively. We firstly demonstrated an erbium-doped fiber laser (EDFL) passively mode-locked by a few-layer $MoS_2$ SA operating at 1.56 μm.[16] Subsequently, Liu *et al.* achieved femtosecond pulses in an EDFL mode-locked with an $MoS_2$-polymer composite SA.[17] These reported results show that few-layer $MoS_2$ can serve as a promising SA for mode-locked fiber lasers.

Currently, there is growing interest in high-repetition-rate ultrafast fiber lasers because of their potential applications in high-speed optical communication,[18] high-resolution frequency metrology,[19] and optical analog-to-digital conversion.[20] The fundamental cavity frequency of a mode-locked fiber ring laser is determined by the cavity length. In order to achieve mode-locked pulses with high repetition rate, it is necessary to reduce the cavity length as short as possible. However, in practice, a minimal cavity length is strongly limited by the pigtail fiber length needed for splicing optical components in a laser cavity. Passive harmonic mode-locking (HML) is an alternative method for multiplying the repetition rate of mode-locked pulses in a fiber laser without reducing the cavity length to impractical levels or requiring any active modulators.[21] Several experimental observations on the passive HML have been reported in fiber lasers mode-locked by NPR technique,[22] SWNTs,[23] graphene,[24] and topological insulator (TI)-based SAs.[25,26] Very recently, Liu *et al.* demonstrated the generation of HML in a fiber laser using a microfiber-based $MoS_2$ SA.[27] The laser produced picosecond pulses at 1558 nm with a repetition rate up to 2.5 GHz. However, the output characteristics of HML, such as stability of pulses, have not been fully addressed. It should

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be noted that the $MoS_2$ SAs used in Refs. [13]– [15], [17], and [27] were prepared by liquid-phase exfoliation (LPE), which contain $MoS_2$ nano-flakes with different layers. It indicates that the prepared $MoS_2$ films are not always uniform. The chemical vapor deposition (CVD) technique can be used to grow high-quality $MoS_2$ films with a precisely controlled number of layers. However, to date, the CVD-grown few-layer $MoS_2$ as an SA for HML in a fiber laser has not yet been reported.

In this paper, we demonstrate a harmonically mode-locked EDFL based on a few-layer $MoS_2$ SA. The pulse repetition rate could be scalable from 10.79 MHz (fundamental cavity frequency) to 216 MHz (20th harmonic) by simply increasing the pump power. The pulse duration is 1.42 ps and the side-mode suppression ratio (SMSR) is ∼36.1 dB at the 20th HML.

## 2. Experimental setup

The few-layer $MoS_2$ used in this study was synthesized in a tube furnace by the CVD method.[28] $MoO_3$ powder was placed in the center of the furnace and sulfur powder was placed at the upstream. During the growth, the pressure inside the tube reactor was kept stable at 0.1 Torr (1 Torr = $1.33322 \times 10^2$ Pa) with constant Ar flow gas. Sulfur powder was heated by a heating belt and the temperature was kept at 100 °C. The furnace temperature was ramped up to 550 °C and kept constant for 30 min. The $MoS_2$ film was grown on an Si substrate capped with a 300-nm thermal $SiO_2$ layer. With a transfer procedure,[16] the as-grown $MoS_2$ film was transferred and attached the end-face of a fiber connector (FC) to form the few-layer $MoS_2$ SA. The Raman spectrum of the as-grown $MoS_2$ film was measured using a Renishaw 100 Raman spectrometer with 514-nm laser. As shown in Fig. 1, the $MoS_2$ film exhibited two Raman characteristic bands at 404.0 $cm^{-1}$ and 379.8 $cm^{-1}$, corresponding to the $A_{1g}$ and $E^1_{2g}$ modes, respectively.[29] The peak frequency difference ($\Delta$) between the $A_{1g}$ and $E^1_{2g}$ modes can be used to identify the number of $MoS_2$ layers.[29] In our case, the $\Delta$ value of the as-grown sample was 24.2 $cm^{-1}$, corresponding to a layer number of 4∼5. Details regarding the characterization of the few-layer $MoS_2$ SA were reported in Ref. [16]. Our experimental results showed that the CVD-grown few-layer $MoS_2$ possessed large saturable absorption at 1550 nm.[16]

The experimental setup of the proposed EDFL is shown in Fig. 2. The ring cavity consists of 0.8-m-long erbium-doped fiber (EDF, Liekki Er80-8/125) with group velocity dispersion (GVD) parameter of $-20$ $ps^2$/km and 18.2-m-long single-mode fiber (SMF) with GVD parameter of $-23$ $ps^2$/km. The net cavity dispersion is estimated to be about $-0.44$ $ps^2$. The laser was pumped by a 500-mW/980-nm laser diode (LD) through a wavelength division multiplexer (WDM), and the laser emission was output through a 10% fiber coupler. A polarization controller (PC) was used to adjust polarization for mode-locking optimization. A polarization-insensitive isolator (ISO) was inserted into the cavity to ensure the unidirectional operation. The FC coated with $MoS_2$ film (see the inset of Fig. 2) was used for mode locking. It should be emphasized that there was no polarization sensitive component used in the laser cavity. Therefore, the effect of mode-locking by nonlinear polarization rotation (NPR) was eliminated. The laser output performance was observed using a 10-GHz photodetector together with a 500-MHz oscilloscope (Tektronix, TDS3052C), a radio-frequency (RF) spectrum analyzer (Advantest, R3267), an optical spectrum analyzer (Yokogawa, AQ 6370C) and an autocorrelator.

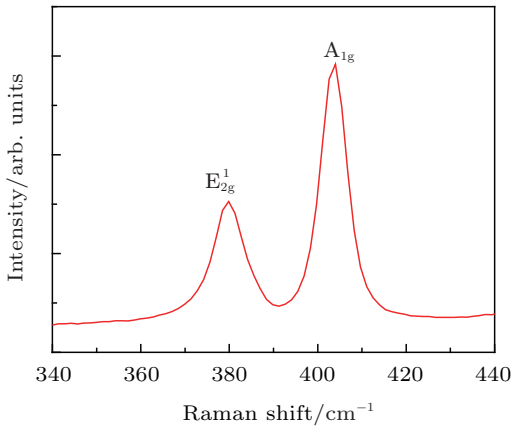



**Fig. 1.** (color online) Raman spectrum of as-grown few-layer $MoS_2$.

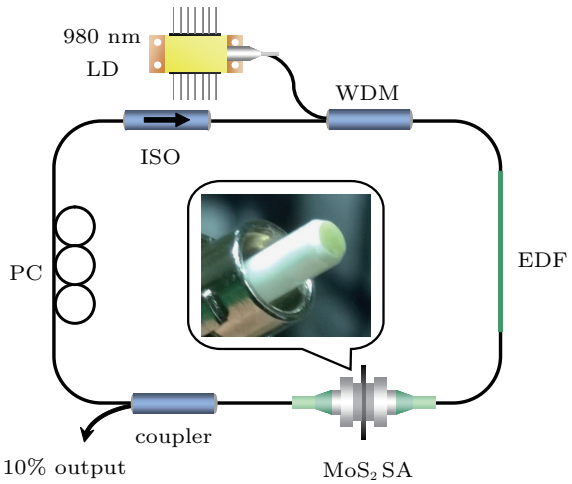



**Fig. 2.** (color online) Experimental setup of the $MoS_2$ mode-locked fiber ring laser. Inset: photograph of an FC coated with few-layer $MoS_2$.

## 3. Experimental results and discussion

Continuous wave (CW) operation of the proposed laser started at a pump power of ∼23 mW. When the pump power was increased to ∼32 mW, the self-starting mode locking was achieved with appropriate adjustment on the PC. Figure 3 shows the laser performance of the fundamental mode-locking

The recorded oscilloscope trace of the mode-locked pulses is shown in Fig. 3(a), which gives a cavity roundtrip time of 92.6 ns. The corresponding radio-frequency (RF) spectrum with a 3-kHz resolution bandwidth is presented in Fig. 3(b). The fundamental repetition rate is 10.79 MHz, matching the cavity roundtrip time of 92.6 ns and cavity length of ∼19 m. The signal-to-noise ratio (SNR) is ∼ 60.2 dB, indicating good mode-locking stability. Figure 3(c) shows the optical spectrum of output pulses, which has a central wavelength of 1569.6 nm and a 3-dB spectral width of approximately 2.4 nm. The Kelly sidebands appear in the optical spectrum, confirming the soliton feature of the mode-locked pulses. A continuous wave (CW) component can be found near the central wavelength, as typically observed in certain fiber lasers mode-locked with TI:$Bi_2Te_3$ SA,[25] or SWNTs.[30] Figure 3(d) presents the pulse autocorrelation trace. The pulse duration is estimated to be 1.17 ps by assuming a $\text{sech}^2$ pulse profile. The time bandwidth product (TBP) of the mode-locked pulses is calculated to be 0.35. The small deviation from the TBP value of 0.315 expected for transform-limited $\text{sech}^2$ pulses indicates that the mode-locked pulses are slightly chirped. To verify that the passive mode-locking was attributed to the few-layer $MoS_2$ SA, the FC coated with $MoS_2$ was replaced with a common clean FC. In this case, no mode-locked pulses were observed even when the PC was rotated and the pump power was adjusted in a wide range. This finding clearly confirmed that the $MoS_2$ SA was responsible for the passive mode-locking operation of the proposed EDFL.

At a pump power greater than 44 mW, pulse splitting and multi-pulsing operations were observed. With proper adjustment on the PC, the pulses equally located in the cavity and the HML operation was observed. Higher-order HML operation was achieved just by carefully increasing the pump power (without adjusting the PC). Figure 4(a) illustrates typical oscilloscope traces of the 5th, 11th, and 18th HML pulses. It was experimentally found that the harmonic order (corresponding to the pulse repetition rate) strongly depended on the pump power level, which was a typical characteristic of the passively HML in fiber lasers.[24–27] Figure 4(b) displays the harmonic order and average output power as functions of the incident pump power. By increasing the pump power from 44 mW to 223 mW, the harmonic order increased from the 2nd to 20th (corresponding to the pulse repetition rates from 21.6 MHz to 216 MHz), and the output power ranged from 0.79 mW to 6.81 mW. The order of HML as well as output power increased almost linearly with the pump power. The HML can be reproduced again next time when the LD pumping source was restarted. On the other hand, after HML operation was obtained, the order of HML was reduced by decreasing the pump power The pump power hysteresis effect of HML operation was found Similar phenomena were also observed in multiple-soliton generation from passively mode-locked fiber laser.[31]

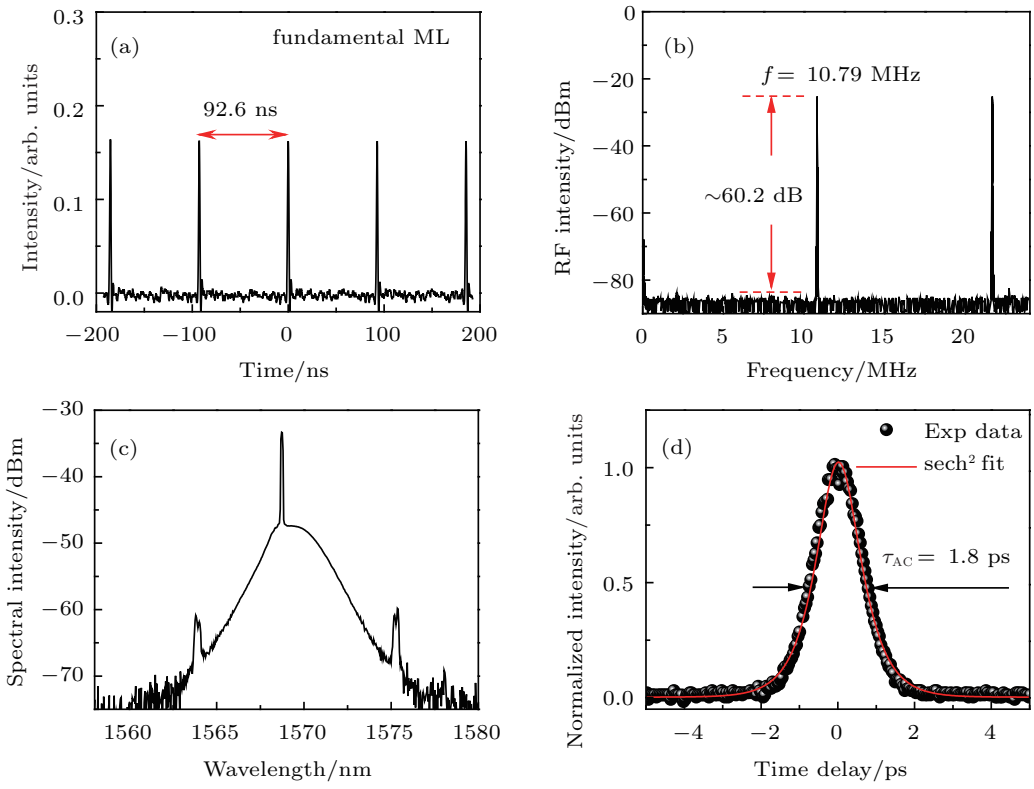



**Fig. 3.** (color online) Performance of the laser mode-locked with the fundamental repetition rate. (a) Output pulse train; (b) RF spectrum measured at 3-kHz RBW, presenting the SNR at a level of 60.2 dB; (c) Optical spectrum; (d) Autocorrelation trace.

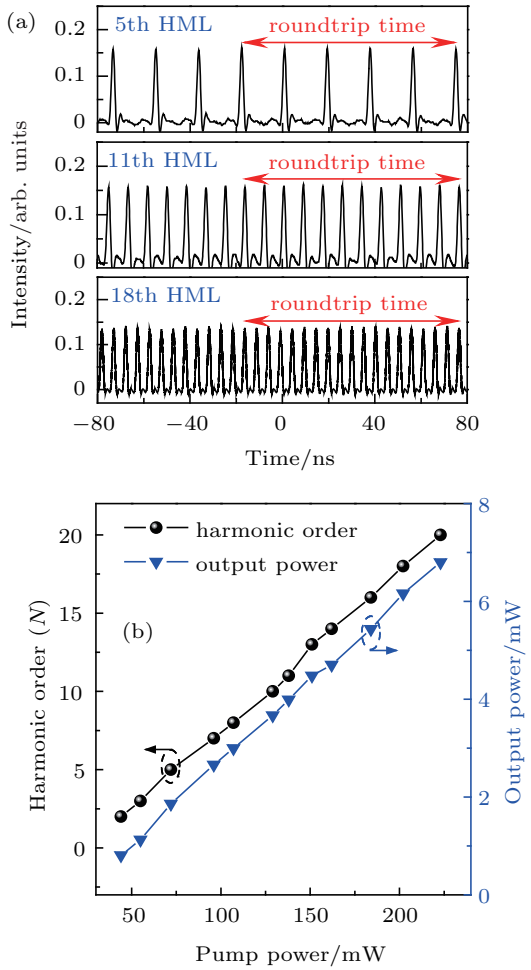



**Fig. 4.** (color online) (a) Output pulse trains of 5th, 11th, and 18th HML; (b) Harmonic order and average output power versus pump power.

Figure 5 illustrates the laser performance of the 20th HML at a pump power of 223 mW. A recorded oscilloscope trace of output pulses is shown in Fig. 5(a). The time interval between the HML pulses is approximately 4.63 ns. The corresponding RF spectrum is shown in Fig. 5(b). The SMSR of the 20th HML was estimated to be approximately 36.1 dB. The corresponding repetition rate was 216 MHz, which agreed well with the 20th harmonic of the fundamental cavity frequency. The optical spectrum of the 20th HML was centered at 1570.3 nm with a 3-dB bandwidth of 2.8 nm, as shown in Fig. 5(c). The autocorrelation trace of output pulses is shown in Fig. 5(d), which gives a pulse duration of 1.42 ps if a $\text{sech}^2$ pulse profile is assumed. The TBP is calculated to be 0.48, which is larger than that of fundamental mode-locked pulses. Experimentally, we found that the higher the harmonic order, the larger the TBP value. When the pump power was increased over 223 mW, the HML operation disappeared due to the unsynchronization between longitudinal modes[26] and the laser started to operate in the CW regime. Similar phenomenon was also observed in a harmonically mode-locked EDFL based on an $Sb_2Te_3$ TI SA.[26]

It should be noted that, a CW component also exists in the 20th harmonic optical spectrum, as shown in Fig. 5(c), which implies the CW emission coexisting with the HML operation. Actually, coexistence of CW lasing and HML operation was also observed in harmonically mode-locked fiber lasers based on NPR technique[22] or graphene saturable

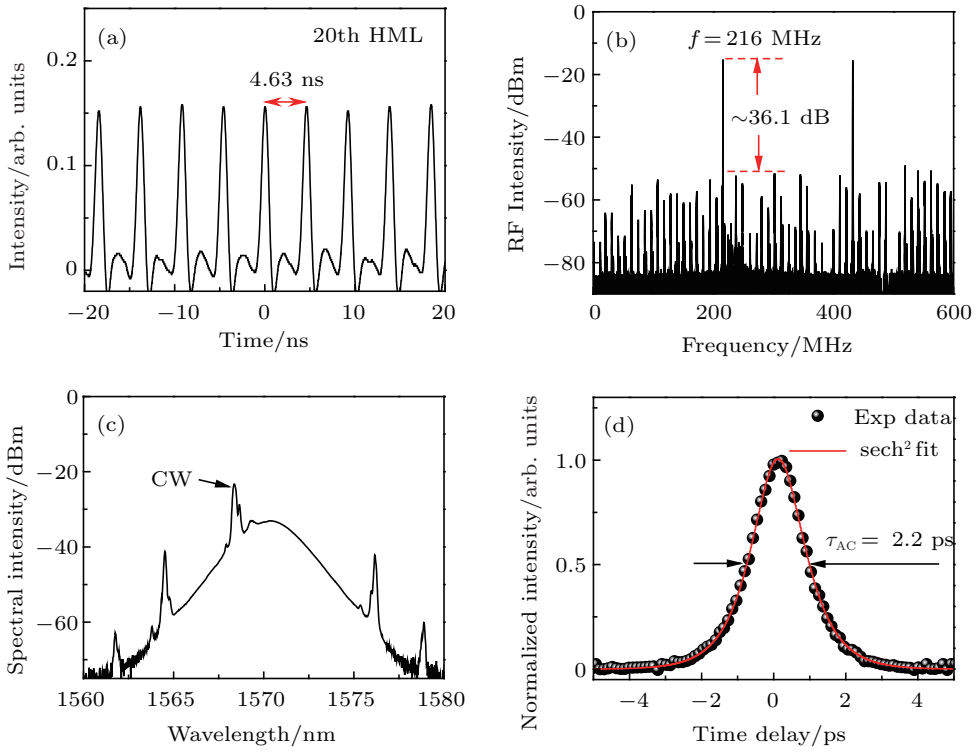



**Fig. 5.** (color online) Performance of the laser operating at the 20th HML. (a) Output pulse train; (b) RF spectrum measured at 100-kHz RBW, presenting the SMSR at a level of 36.1 dB; (c) Optical spectrum; (d) Autocorrelation trace.

absorber.[32] When the CW component was settled in proper position in pulse spectrum by carefully adjusting the PC, the phases between multiple pulses were synchronized and the pulses equally located in the laser cavity. As a result, HML operation appeared. It is suggested that the CW component as an efficient mediator plays an important role for stabilizing the HML operation of the laser.[32,33] In our experiment, to test the long-term stability of the 20th HML, the laser was turned on over two hours in the conventional laboratory condition. Relative fluctuations of average output power and center wavelength were about $\pm 3\%$ and $\pm 0.15\%$, respectively, indicating the good stability of the 20th HML in the room environment. It should be emphasized that the parameters of output pulses generated by a harmonically mode-locked fiber laser, such as the highest order of HML, pulse duration and SMSR, are strongly dependent on the cavity dispersion map and the SA performance.[23,30] We believe that multi-GHz pulse repetition rate in our harmonically mode-locked fiber laser can be achieved by optimizing the SA parameters of few-layer $MoS_2$ and the cavity design.

## 4. Conclusion

We have demonstrated a harmonically mode-locked EDFL based on a few-layer $MoS_2$ SA The few-layer $MoS_2$ film was synthesized by the CVD method. The 20th HML of the fiber laser was achieved at a repetition rate of 216 MHz with a pulse duration of 1.42 ps and SMSR of 36.1 dB. Further efforts would be made to obtain multi-GHz repetition rate in HML fiber lasers for practical applications, such as high-speed optical communication and optical analog-to-digital conversion.

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