

# Investigation of nonlinear optical properties of rhenium diselenide and its application as a femtosecond mode-locker

JINHO LEE,  KYUNGTAEK LEE,  SUHYOUNG KWON, BUMSOO SHIN, AND JU HAN LEE\* 

School of Electrical and Computer Engineering, University of Seoul, Seoul 02504, South Korea

\*Corresponding author: j.h.lee@ieee.org

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We investigated the nonlinear optical properties of ReSe<sub>2</sub>. First, we measured the nonlinear absorption coefficient and the nonlinear refractive index of a ReSe<sub>2</sub> thin film using open-aperture (OA) and closed-aperture (CA) Z-scan techniques. ReSe<sub>2</sub> was shown to possess both saturable absorption and self-defocusing properties. The nonlinear absorption coefficient of ReSe<sub>2</sub> was measured to be  $-(5.67 \pm 0.35) \times 10^3$  cm/GW, and its nonlinear refractive index was  $-(2.81 \pm 0.13) \times 10^{-2}$  cm<sup>2</sup>/GW at 1560 nm. Next, a fiberized saturable absorber (SA) based on ReSe<sub>2</sub> was fabricated with a side-polished fiber platform and was tested as an ultrafast mode-locker capable of producing femtosecond pulses operating at a wavelength of 1560 nm. The estimated modulation depth and saturation power are  $\sim 3.9\%$  and  $\sim 42$  W, respectively, for the transverse electric mode, while they are  $\sim 2.4\%$  and  $\sim 53$  W for the transverse magnetic mode. Using the prepared SA, stable soliton pulses with a temporal width of  $\sim 862$  fs were produced from an erbium-doped fiber ring cavity. To the best of the authors' knowledge, this is the first demonstration of using a ReSe<sub>2</sub>-based SA for femtosecond mode-locked pulse generation. © 2019 Chinese Laser Press

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## 1. INTRODUCTION

Optical materials with nonlinear properties have always been important for the field of nonlinear optics (NLO) since they can be used for practical applications such as optical switching, mode-locking, and optical limiting [1–9]. Optical nonlinearity means that the physical parameters of a material, such as absorptivity and refractive index, are electric field intensity dependent. In the past several decades, a number of materials have been found to possess significant nonlinear optical properties [10–12]. In particular, nanostructured materials have recently been a focus of research since they provide a flexible platform for the implementation of various functional electronic or photonic devices that are difficult to realize with traditional bulk-structured material platforms. Nanostructured nonlinear optical materials include carbon nanotubes (CNTs) [13,14], graphene [15–17], graphene oxide (GO) [18,19], topological insulators (TIs) [20–25], transition-metal dichalcogenides (TMDCs) [6,8,26–45], black phosphorus (BPs) [46–48], gold nanoparticles [49,50], skutterudites [51,52], MXenes [53,54], and transition-metal monochalcogenides (TMMCs) [55]. It should be noted that nanostructured materials do not imply two-dimensional (2D) materials even if 2D materials are included in the material structure.

One family of recently discovered nanostructured materials consists of TMDCs [56]. TMDCs are materials with a formula

of  $MX_2$ , where  $M$  is a transition metal element (e.g., Mo, W, and Nb) and  $X$  is a chalcogen (e.g., S, Se, or Te). TMDCs have a layered  $X-M-X$  structure, wherein a layer of the transition metal atom is sandwiched between two layers of chalcogen atoms. The adjacent layers are weakly bonded together by van der Waals forces, and therefore thin sheets can be easily exfoliated from the bulk material. A number of investigations of the nonlinear optical properties of TMDCs have been conducted thus far, and their utilization in optical devices has been extensively demonstrated. For example, Zhang *et al.* reported the saturable absorption property of molybdenum disulfide (MoS<sub>2</sub>) for the first time [6], to the best of our knowledge. Two-photon absorption (TPA) of tungsten disulfide (WS<sub>2</sub>) and MoS<sub>2</sub> was investigated by Zhang *et al.* [57]. The second and third-order optical susceptibilities of MoS<sub>2</sub> were investigated by a number of research groups [58–60].

Rhenium diselenide (ReSe<sub>2</sub>) is a rhenium-based TMDC. Compared to commonly known molybdenum- or tungsten-based TMDCs, rhenium-based TMDCs such as ReS<sub>2</sub> and ReSe<sub>2</sub> have attracted less technical attention until recently, even though they possess electronic or optical properties comparable to the aforementioned common TMDCs [61–63]. Very recently, quite a few investigations of the nonlinear optical properties of ReS<sub>2</sub> and ReSe<sub>2</sub> have been conducted [34–42]. In particular, most of the investigations of ReSe<sub>2</sub> have focused

on saturable absorption. For example, Du *et al.* reported saturable absorption properties of ReSe<sub>2</sub> for the first time, to the best of our knowledge, and demonstrated a ReSe<sub>2</sub>-based saturable absorber (SA) for a Q-switched fiber laser [39]. Li *et al.* systematically investigated nonlinear saturable absorption properties of ReSe<sub>2</sub> at 1030 and 515 nm using an open-aperture (OA) Z-scan technique and demonstrated its use for picosecond mode-locking of a 1064 nm waveguide laser [40]. Investigations regarding ReSe<sub>2</sub>-based SAs have also been conducted by Li *et al.* [41,42].

In this paper, we report a detailed investigation of the nonlinear optical properties of ReSe<sub>2</sub> at a wavelength of 1560 nm. This investigation was conducted with three-dimensional (3D) structured ReSe<sub>2</sub>, as opposed to 2D structured. First, we show experimental measurements of the nonlinear absorption coefficient and the nonlinear refractive index of a thin ReSe<sub>2</sub> film with an OA and closed-aperture (CA) Z-scan technique. Note that unlike our measurements, the Z-scan measurement of ReSe<sub>2</sub> by Li *et al.* was based on an OA technique to obtain limited information about saturable absorption properties at 1030 and 515 nm [40]. Our measurements are conducted with a femtosecond fiber laser at a wavelength of 1560 nm and show that ReSe<sub>2</sub> has both saturable absorption and self-defocusing properties. Next, a fiberized SA based on ReSe<sub>2</sub> is fabricated with a side-polished fiber platform. Finally, the fabricated SA is tested as an ultrafast mode-locker and was found to be capable of producing femtosecond pulses operating at a wavelength of 1560 nm. To the best of the author's knowledge, there have been no previous reports of ReSe<sub>2</sub>-based SAs for use in the femtosecond mode-locking of a laser.

## 2. FABRICATION AND CHARACTERIZATION OF A ReSe<sub>2</sub>-BASED THIN FILM

To fabricate a ReSe<sub>2</sub> thin film, ReSe<sub>2</sub> bulk flakes (99.99%, HQ graphene) were used as a starting material. Nanometer- or micrometer-sized ReSe<sub>2</sub> particles were first obtained using bath-type ultrasonication in 10 mL of distilled water for 16 h. Note that no additional centrifugation step for elimination of large agglomerates was conducted in our case, since our target particles were not nano-sized particles. Figure 1(a) shows scanning electron microscopy (SEM) image of the obtained

ReSe<sub>2</sub> particles. Their sizes are observed to vary from tens of nanometers to around 2 μm. An energy-dispersive spectroscopy (EDS) measurement was also conducted, as shown in Fig. 1(b). For the EDS measurement, a small amount of solution containing ReSe<sub>2</sub> particles was dropped and dried on top of a glass slide. The two strong peaks observed at 1.85 keV and 1.4 keV are associated with Re (M<sub>α</sub>) and Se (L<sub>α</sub>) [64], and the atomic ratio between Re and Se was approximately 1:2.

To form a thin film, polyvinyl alcohol (PVA) was mixed with the prepared ReSe<sub>2</sub> solution. The concentration of ReSe<sub>2</sub> in the ReSe<sub>2</sub>/PVA composite was ~1 mg · mL<sup>-1</sup>. An atomic force microscope (AFM) image of our prepared ReSe<sub>2</sub> thin film has been measured, as shown in Fig. 2. The thickness of the prepared ReSe<sub>2</sub> particles within the target range was measured to be ~450 nm. However, note that the thickness of prepared ReSe<sub>2</sub> particles was random since no centrifugation process was used after ultrasonication. It was thus impossible to obtain a reasonable average thickness and layer number from a limited number of AFM-measured values.

To analyze the stoichiometry of the ReSe<sub>2</sub>/PVA thin film, X-ray photoelectron spectroscopy (XPS) measurements were performed. High-resolution spectra of the Re 4f and Se 3d region are shown in Figs. 3(a) and 3(b), respectively. The two peaks of Re 4f<sub>7/2</sub> and Re 4f<sub>5/2</sub> at ~41.6 and ~44 eV are consistent with previously reported results [64,65]. Two peaks at 54.6 and ~55.6 eV in the Se-3d region of Fig. 3(b) are consistent with the peaks of Se 3d<sub>5/2</sub> and Se 3d<sub>3/2</sub> [64,65]. The measured high-resolution spectrum for the C 1s region confirms the existence of CH<sub>2</sub> and CH-OH groups in the PVA [66]. The peaks centered at the binding energies of ~284 eV, ~286 eV, ~288.7 eV in the C 1s region can be attributed to CH<sub>2</sub>, CH-OH, and C=O groups, respectively [66,67].

Next, we measured the Raman spectrum of the ReSe<sub>2</sub>/PVA thin film at 532 nm (InVia Raman Microscope, Renishaw) [Fig. 4(a)]. Many small peaks were observed due to the complex lattice vibration and low symmetry of ReSe<sub>2</sub> used [41,42,64,65,68–70]. Three main Raman peaks at ~124, ~159, and ~173 cm<sup>-1</sup> indicate in-plane (E<sub>g</sub>) and out-of-plane (A<sub>g</sub>) vibrational modes [41,42,64,65,68–70]. Figure 4(b) shows the measured linear optical absorption of the ReSe<sub>2</sub>/PVA composite film over a spectral range of 1000–1800 nm, which was measured using a spectrophotometer

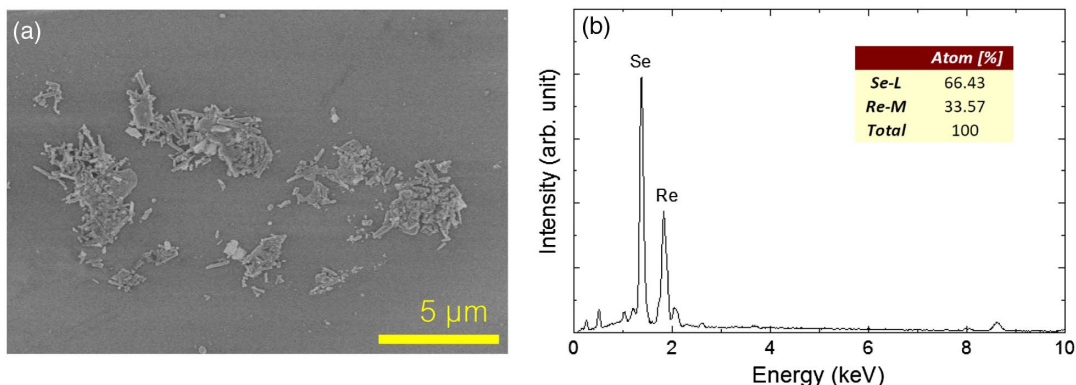
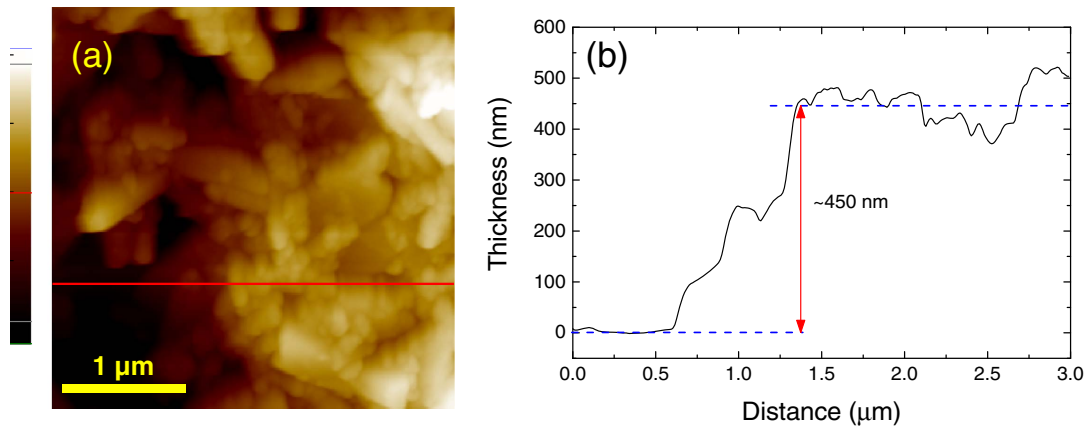
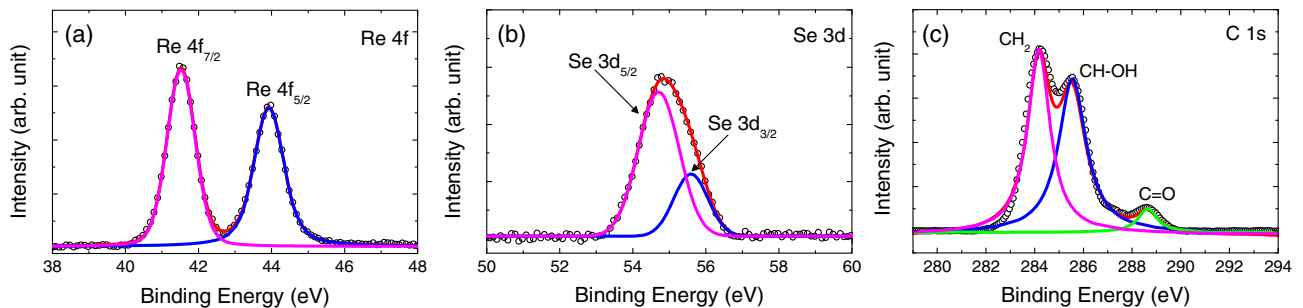


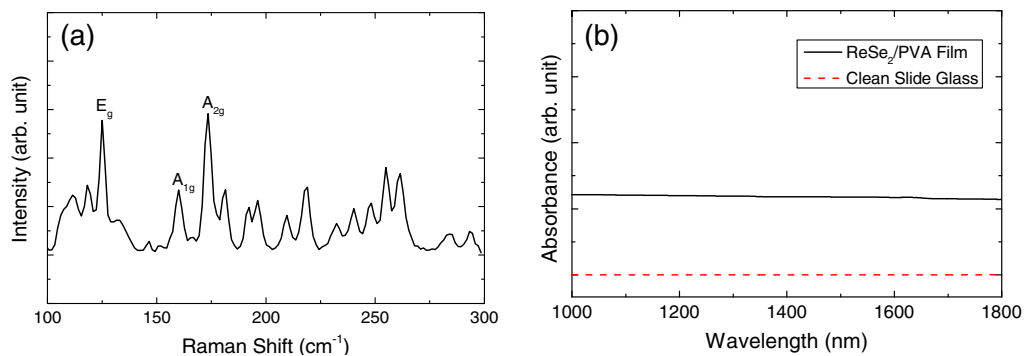
Fig. 1. (a) SEM image and (b) EDS spectrum of the prepared ReSe<sub>2</sub> particles.



**Fig. 2.** (a) AFM image and (b) line profile of the ReSe<sub>2</sub> thin film.



**Fig. 3.** XPS spectra of the prepared ReSe<sub>2</sub>/PVA thin film in the (a) Re 4f region, (b) Se 3d region, and (c) C 1s region.



**Fig. 4.** (a) Measured Raman spectrum of the ReSe<sub>2</sub>/PVA thin film. (b) Linear optical absorption spectrum of the ReSe<sub>2</sub>/PVA composite.

(UV-3600PLUS, Shimadzu). For the measurement, the ReSe<sub>2</sub>/PVA composite solution was deposited onto a glass slide. A broad absorption band of the prepared ReSe<sub>2</sub>/PVA composite was clearly observed, which included absorption in the 1.5  $\mu\text{m}$  wavelength band.

It was reported that the energy bandgap of bulk ReSe<sub>2</sub> is around 0.995 eV (1246 nm), while that of a ReSe<sub>2</sub> monolayer is 1.24 eV (1000 nm) [70,71]. From the perspective of the energy bandgap, it should thus be impossible to observe photon

absorption from the ReSe<sub>2</sub> particles at wavelengths above 1246 nm. However, as shown in Fig. 4(b), the optical absorption of the prepared ReSe<sub>2</sub>/PVA film was clearly observed in the 1.5  $\mu\text{m}$  wavelength region. Such absorption behavior could be attributed to Re and Se atomic defects [40]. As mentioned in Ref. [40], the atomic defects could reduce the energy bandgap of ReSe<sub>2</sub>, allowing for broadband photon absorption irrespective of the energy bandgap of the pristine material. In fact, sub-bandgap absorption has been previously reported in other

TMDC materials. It is well-known that the broken symmetry and unsatisfied bonds between  $M$  and  $X$  atoms at edges can modify the electronic structures of TMDCs [72].

### 3. Z-SCAN MEASUREMENT FOR A ReSe<sub>2</sub>-BASED THIN FILM

The measurements for the nonlinear optical response of the ReSe<sub>2</sub> particles were carried out by using the OA and CA Z-scan techniques [73]. The experimental setup is shown in Fig. 5(a). A homemade mode-locked fiber laser with a repetition rate of 22.26 MHz and a pulse width of 300 fs at a wavelength of 1560 nm was used as an input source. To obtain the nonlinear absorption coefficient, an OA Z-scan measurement was first performed with the aperture fully opened ( $S = 1$ ). The beam was focused through a plano-convex lens to obtain a peak intensity of about 50 MW/cm<sup>2</sup>. The position of the sample was changed from  $z = -15$  mm to  $z = 15$  mm with a step size of 0.5 mm. The spot size was estimated from the obtained curve, which was approximately 22  $\mu\text{m}$  at the focal point. The average thickness of the prepared ReSe<sub>2</sub>/PVA film was approximately 9  $\mu\text{m}$ . The formed ReSe<sub>2</sub>/PVA thin film had a circular shape with a diameter of  $\sim 10$  mm, which is much larger than the beam waist of  $\sim 22$   $\mu\text{m}$ . The Z-scan setup was calibrated considering the effect of the substrate.

As shown in Fig. 5(b), the change of the transmittance of the sample was measured with a photodiode sensor; the normalized values of the measured data were plotted against the  $z$  position and fitted with the following formula [73,74]:

$$T(z) = \sum_{n=0}^{\infty} (-\beta I_0 L_{\text{eff}})^n / (1 + z^2/z_0^2)^n (n+1)^{3/2} \approx 1 - \beta I_0 L_{\text{eff}} / 2^{3/2} (1 + z^2/z_0^2), \quad (1)$$

where  $T(z)$  is the normalized transmittance,  $\beta$  is the nonlinear absorption coefficient,  $I_0$  is the peak on-axis intensity at the focus,  $L_{\text{eff}}$  is the effective length,  $z$  is the position of the sample, and  $z_0$  is the Rayleigh length.

To determine the sign and magnitude of the nonlinear refractive index of the ReSe<sub>2</sub> sample, a CA Z-scan measurement was subsequently conducted, while the aperture at the far field was kept closed ( $S \sim 0.7$ ). In the CA Z-scan measurement, the transmitted beam is sensitive to both nonlinear absorption and nonlinear refraction. By dividing the normalized results of a CA Z scan by those of an OA Z scan, we can distinguish the

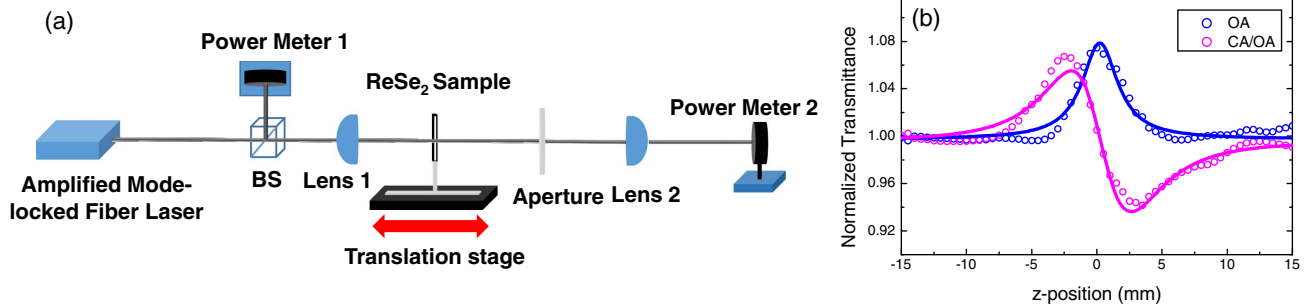
nonlinear refraction from the nonlinear absorption [73]. The normalized values of the measured data were plotted against the  $z$  position and fitted with the following formula [75]:

$$T(z) = \frac{1}{1 - \frac{4z/z_0}{(1+z^2/z_0^2)^2} \Delta\Phi + \frac{4}{(1+z^2/z_0^2)^3} \Delta\Phi^2}, \quad (2)$$

where  $\Delta\Phi = 2\pi n_2 I_0 L_{\text{eff}} / \lambda$  is the phase change,  $n_2$  is the nonlinear refractive index,  $L_{\text{eff}} = (1 - e^{-\alpha_0 L}) / \alpha_0$  is the effective thickness of the sample,  $\alpha_0$  is the linear absorption coefficient,  $L$  is the thickness of the sample,  $z_0 = \pi \omega_0^2 / \lambda$  is the Rayleigh length,  $\omega_0$  is the beam waist,  $\lambda$  is the wavelength, and  $I_0$  is the intensity at the focusing point. The measured nonlinear absorption coefficient of the ReSe<sub>2</sub>/PVA film was  $-(5.67 \pm 0.35) \times 10^3$  cm/GW, and its nonlinear refractive index was  $-(2.81 \pm 0.13) \times 10^{-2}$  cm<sup>2</sup>/GW.

The curves in Fig. 5(b) show the nonlinear saturable absorption and self-defocusing behaviors of the ReSe<sub>2</sub> film at a wavelength of 1560 nm. Considering the bandgap of the pristine ReSe<sub>2</sub> [70,71], the ReSe<sub>2</sub> was supposed to exhibit TPA at 1560 nm; however, no TPA was observed in our ReSe<sub>2</sub> sample [76,77]. On the other hand, our measurement results revealed that ReSe<sub>2</sub> possesses a saturable absorption property, even for incident photons with energies lower than its bulk bandgap energy. The nonlinear saturable absorption of ReSe<sub>2</sub> could be attributed to a defect-driven bandgap decrease or edge state absorption [72]. Note that TMDC materials with bandgap energies larger than the incident photon energy are known to exhibit such a saturable absorption behavior in the 1550 nm wavelength region [76,77]. Table 1 compares the Z-scan-measured nonlinear optical parameters of a variety of TMDCs with those of ReSe<sub>2</sub>. Note that the previous measurements for nonlinear absorption coefficients and the nonlinear refractive indices of TMDCs were conducted at wavelengths of 515, 800, 1030, or 1064 nm. Since our measurements were conducted at 1560 nm, it is impossible to directly compare our measured nonlinear optical parameters of ReSe<sub>2</sub> to those of the other MDCs. It is believed that ReSe<sub>2</sub> has nonlinear optical properties comparable to various TMDCs.

In this experiment, we have not observed TPA in our prepared ReSe<sub>2</sub>/PVA film. This could be attributed to the fact that our used pulse laser for the Z-scan measurements might not have a peak power high enough to induce TPA. Regarding the effect of a large nonlinear absorption coefficient  $\beta$  on the mode-locking performance, we believe that a large nonlinear



**Fig. 5.** (a) Schematic diagram of the Z-scan measurement setup at 1560 nm. BS: beam splitter. (b) Z-scan results for the ReSe<sub>2</sub> film using open-aperture (OA) and closed-aperture (CA) Z scan.

**Table 1. Comparison of Nonlinear Optical Parameters for a Range of TMDCs that Were Measured with the Z-Scan Technique**

Sample	NLO Response	$\lambda$ (nm)	$\alpha_0$ (cm <sup>-1</sup> )	$\beta$ (cm/GW)	$n_2$ (cm <sup>2</sup> /GW)	Reference
WS <sub>2</sub>	SA <sup>a</sup>	800	8.88 nm <sup>-1</sup>	$-(3.7 \pm 0.28) \times 10^5$	$8.1 \pm 0.41$	[78]
WS <sub>2</sub>	SA	1064	NA <sup>c</sup>	$-(5.1 \pm 0.26)$	$(5.83 \pm 0.18) \times 10^{-2}$	[79]
MoS <sub>2</sub>	SA	1064	NA	$-(3.8 \pm 0.59)$	$(1.88 \pm 0.48) \times 10^{-3}$	[79]
WSe <sub>2</sub>	TPA <sup>b</sup>	1064	NA	$(1.9 \pm 0.57)$	$-(2.4 \pm 1.23)$	[79]
Mo <sub>0.5</sub> W <sub>0.5</sub> S <sub>2</sub>	TPA	1064	NA	$(1.91 \pm 0.78)$	$-(8.73 \pm 1.47) \times 10^{-2}$	[79]
WS <sub>2</sub> (1–3L)	TPA	1030	$7.17 \times 10^5$	$(1.0 \pm 0.8) \times 10^4$	NA	[57]
WS <sub>2</sub> (18–20L)	TPA	1030	$5.98 \times 10^5$	$(3.28 \pm 0.11) \times 10^3$	NA	[57]
WS <sub>2</sub> (39–41L)	TPA	1030	$8.57 \times 10^5$	$(2.75 \pm 0.11) \times 10^3$	NA	[57]
MoS <sub>2</sub> (25–27L)	TPA	1030	$3.90 \times 10^4$	$66 \pm 4$	NA	[57]
MoS <sub>2</sub> (72–74L)	SA	1030	$1.89 \times 10^5$	$-250 \pm 50$	NA	[57]
WS <sub>2</sub> (1–3L)	TPA	800	$1.08 \times 10^6$	$525 \pm 205$	NA	[57]
WS <sub>2</sub> (18–20L)	SA	800	$7.22 \times 10^5$	$-397 \pm 40$	NA	[57]
MoS <sub>2</sub> (25–27L)	TPA	800	$6.24 \times 10^4$	$11.4 \pm 4.3$	NA	[57]
WS <sub>2</sub> (1–3L)	SA	515	$5.18 \times 10^4$	$-(2.9 \pm 1.0) \times 10^4$	NA	[57]
ReSe <sub>2</sub>	SA	1560	$0.25 \times 10^3$	$-(5.67 \pm 0.35) \times 10^3$	$-(2.81 \pm 0.13) \times 10^{-2}$	This work

<sup>a</sup>SA, saturable absorption.

<sup>b</sup>TPA, two photon absorption.

<sup>c</sup>NA, not available.

absorption coefficient  $\beta$  of a nonlinear optical material is associated with a large modulation depth of the material-based SA. Note that the use of an SA with a larger modulation depth is known to increase the mode-locking stability as well as to reduce the mode-locked pulse width [80,81]. The relaxation time measurement of the ReSe<sub>2</sub> sample was not conducted since no pump–probe spectroscopy setup was available in our laboratory.

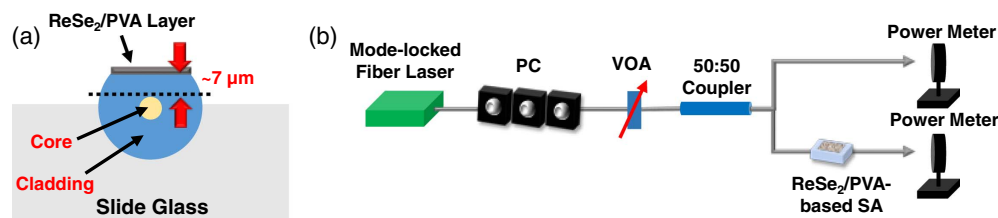
#### 4. FABRICATION OF A ReSe<sub>2</sub>-BASED FIBERIZED SATURABLE ABSORBER

Next, we fabricated a fiberized SA that can be used for ultrafast mode-locking based on ReSe<sub>2</sub>. A side-polished fiber was employed as a platform. The side-polished fiber was prepared through the polishing of one side of an SMF-28 fiber, which was fixed onto the V groove of a glass slide. The cross-sectional structure of the prepared SA is shown in Fig. 6(a). The insertion loss and polarization-dependent loss (PDL) of the prepared side-polished fiber were  $\sim 0.8$  dB and  $\sim 0.05$  dB at 1560 nm, respectively. The physical distance of the flat side to the edge of the fiber core was of  $\sim 7$   $\mu$ m, and the polishing length was  $\sim 2.5$  mm. The ReSe<sub>2</sub>/PVA solution was deposited onto the flat side of the side-polished fiber using the solution-drop method and then dried at room temperature for 24 h. The insertion loss and PDL of the ReSe<sub>2</sub>/PVA-deposited side-polished fiber were  $\sim 2.9$  dB and  $\sim 1.6$  dB, respectively.

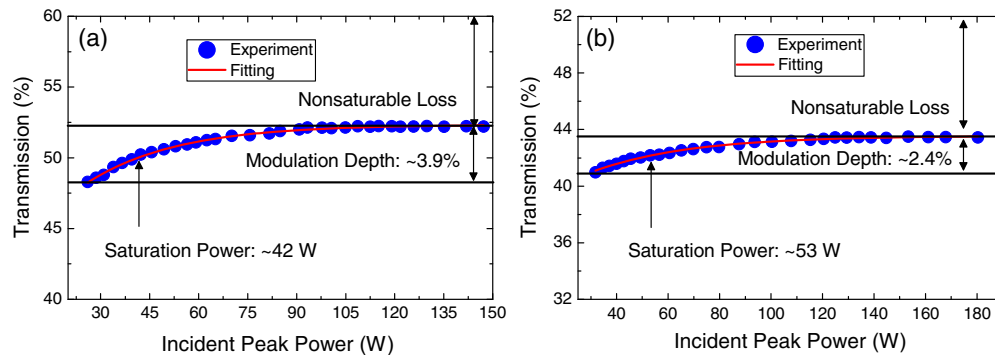
Nonlinear transmission curves of the ReSe<sub>2</sub>/PVA-based SA were measured as a function of the incident peak power of the input pulses for transverse electric (TE) and transverse magnetic (TM) modes, respectively. For this measurement, we used a mode-locked, 1.56  $\mu$ m fiber laser with a temporal width of  $\sim 680$  fs at a repetition rate of  $\sim 22.26$  MHz, and the measurement setup is shown in Fig. 6(b). Since our SA had a non-negligible PDL of  $\sim 1.6$  dB, a polarization controller (PC) was incorporated into the measurement setup. It was straightforward to distinguish the TE and TM modes by measuring the output power while the PC was adjusted. A variable optical attenuator (VOA) was used to adjust the optical power of the mode-locked pulses. A 50:50 coupler was used to split the mode-locked pulses into two ports. One of the two ports was connected to the prepared ReSe<sub>2</sub>/PVA-based SA, while the other was directly connected to a power meter to monitor the input beam power on the prepared SA. Another power meter was used to monitor the output power from the ReSe<sub>2</sub>/PVA-based SA for comparison with the input power. Figure 7 shows the nonlinear transmission curves for the TE and TM modes fitted to the following equation [82]:

$$T(I) = 1 - \Delta T \cdot \exp\left(\frac{-I}{I_{\text{sat}}}\right) - T_{\text{ns}}, \quad (3)$$

where  $T$  is the transmission,  $\Delta T$  is the modulation depth,  $I$  is the incident pulse energy,  $I_{\text{sat}}$  is the saturation energy, and



**Fig. 6.** (a) Schematic of our prepared ReSe<sub>2</sub>/PVA-deposited side-polished fiber. (b) Measurement setup for nonlinear transmission curves of the ReSe<sub>2</sub>/PVA-based SA at 1560 nm.



**Fig. 7.** Nonlinear transmission curves of the ReSe<sub>2</sub>/PVA-deposited side-polished fiber: (a) the transverse electric (TE) mode and (b) the transverse magnetic (TM) mode.

$T_{ns}$  is the nonsaturable loss. The estimated modulation depth and saturation power for the TE mode are  $\sim 3.9\%$  and  $\sim 42$  W, respectively, while they are  $\sim 2.4\%$  and  $\sim 53$  W, respectively, for the TM mode. Due to the asymmetric structure of our prepared ReSe<sub>2</sub>/PVA-deposited side-polished fiber, the TE and TM modes have different interaction strengths between the ReSe<sub>2</sub>/PVA film and oscillating beam. This is the reason why the TE and TM modes exhibit different saturable absorption properties. Regarding the main mode for the actual mode-locking, we believe that the TE mode would play a key role in our laser setup since the TE mode experiences less insertion loss. It is believed that the modulation depth level is high enough to induce soliton mode-locking in the fiberized laser cavity with proper anomalous dispersion [81].

In order to measure the damage threshold of the proposed ReSe<sub>2</sub>/PVA-based SA, we have launched a 1550 nm continuous-wave amplified laser beam of 1 W power into it. However, we have not observed any substantial damage of the device within the optical power level. Therefore, we believe that the damage threshold of the prepared ReSe<sub>2</sub>/PVA-based SA must be larger than 1 W. However, it was impossible to measure the precise value due to the limited availability of a high-power laser in our laboratory.

## 5. MODE-LOCKING PERFORMANCE MEASUREMENTS

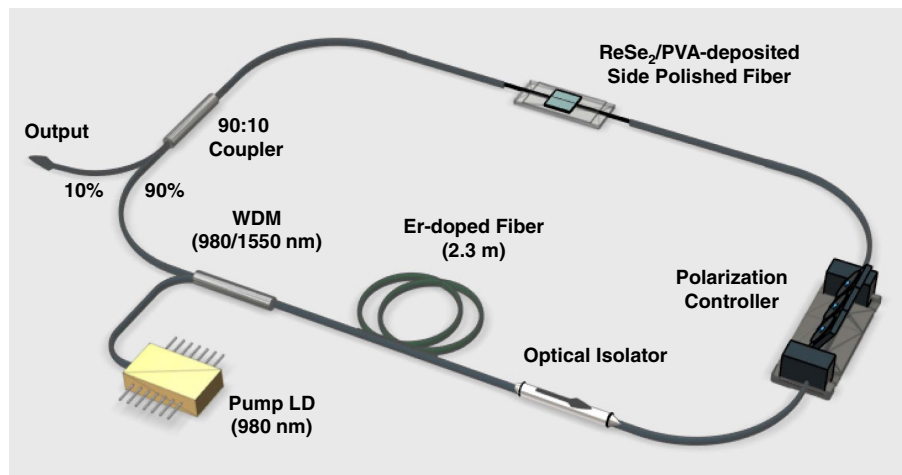
The experimental schematic of our passively mode-locked fiber laser is shown in Fig. 8. The gain medium is a 2.3-m-long erbium-doped fiber (EDF) with peak absorption of  $\sim 20$  dB/m at a wavelength of 1530 nm. A 980 nm pump laser diode was used as the pump source that was inserted into the gain medium through a 980/1550 nm wavelength division multiplexer (WDM). The polarization-independent isolator was used to force unidirectional light propagation. The mode-locked laser output was extracted from the ring cavity using the 10% port of a 90:10 coupler. A PC was used to optimize the polarization state of the oscillating beam within the laser cavity. The ReSe<sub>2</sub>/PVA-deposited side-polished fiber was placed after the PC.

Stable mode-locked pulses were readily obtained when the pump power was set to  $\sim 19.6$  mW, with the PC properly adjusted. The average output power was measured to be

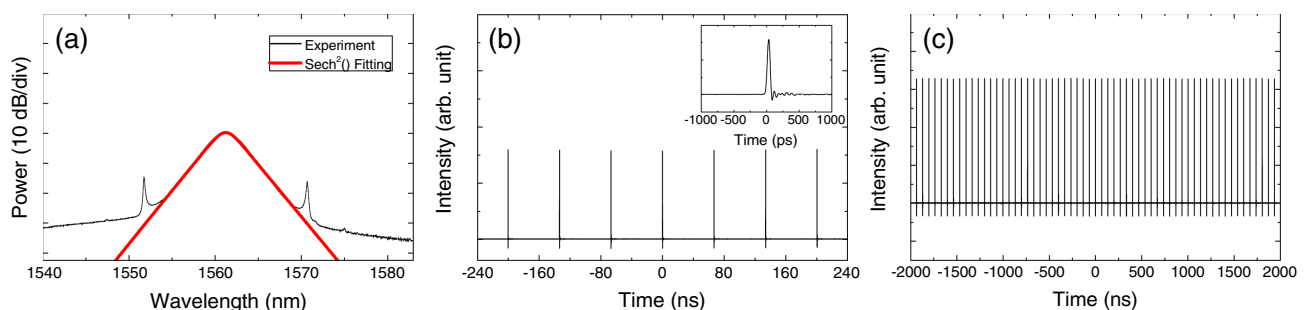
$\sim 0.5$  mW. The main factor limiting the output power is the unoptimized cavity configuration. To optimize the laser cavity to produce higher average output power, we tried to reduce the insertion loss of the prepared SA as well as to change the output coupler. To control the insertion loss of the prepared SA, we need to optimize the side-polished fiber in terms of its polishing depth, interaction length, and the concentration of the solution. Such work to enhance the laser performance is reserved for future work. Figure 9(a) shows the measured optical spectrum of the mode-locked pulses. Kelly sidebands were clearly observed, indicating that the fiber laser was operating in the soliton regime [83]. The center wavelength and 3 dB bandwidth of the pulses were measured to be  $\sim 1561.2$  nm and  $\sim 3.4$  nm, respectively. Figure 9(b) shows the measured oscilloscope trace of the output pulses with a 16 GHz real time oscilloscope combined with an 11 GHz photodetector. The inset of Fig. 9(b) shows a magnified view of a single output pulse. The temporal period of the output pulse is  $\sim 66.8$  ns, which corresponds to the fundamental repetition rate of  $\sim 14.97$  MHz.

Next, an autocorrelation measurement was performed using a TPA-based autocorrelator. Figure 10(a) shows the measured autocorrelation trace of the output pulses with a hyperbolic secant-squared fitting curve. The estimated temporal width of the output pulses is  $\sim 862$  fs. Considering the 3 dB bandwidth of  $\sim 3.4$  nm, the estimated time-bandwidth product is  $\sim 0.361$ , indicating that the output pulses are slightly chirped. The electrical spectrum was measured to confirm the stability of the output pulses, as shown in Fig. 10(b). A strong signal peak with an electrical signal-to-noise ratio (SNR) of  $\sim 65$  dB was obtained at a fundamental repetition rate of  $\sim 14.97$  MHz. The inset of Fig. 10(b) shows the electrical spectrum with a 1 GHz frequency span. Strong harmonic signals were clearly observed, strongly indicating that the output pulses are stable mode-locked pulses.

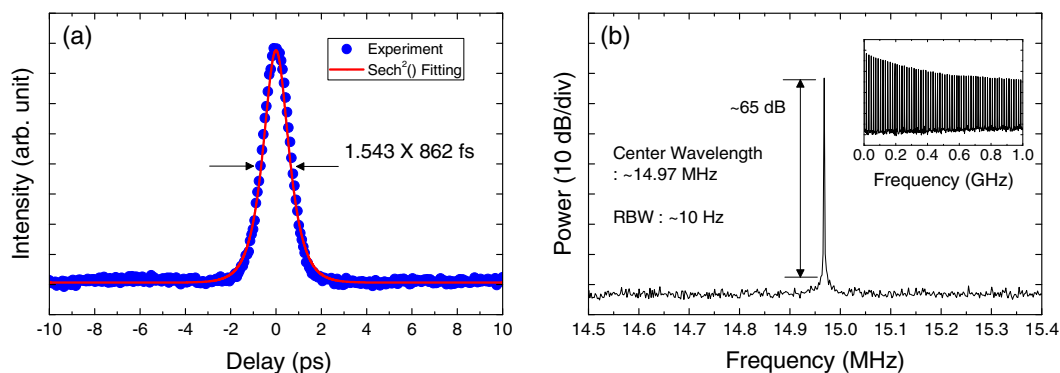
The solution dropping is a simple method to form a thin film on a substrate by dropping and drying a small amount of solution. However, it is difficult to control the uniformity of the film thickness using this method. The saturable absorption performance of our prepared SA was observed to change in some degree sample by sample. However, the output performance of our mode-locked laser was not observed to



**Fig. 8.** Experimental configuration of our mode-locked fiber laser.



**Fig. 9.** (a) Optical spectrum and (b) an oscilloscope trace of the output pulses. Inset: oscilloscope trace for a narrow span. (c) An oscilloscope trace of the output pulses over a large time scale.



**Fig. 10.** (a) Autocorrelation trace and (b) electrical spectrum of the output pulses. Inset: electrical spectrum for a span of 1 GHz.

significantly vary depending on the SA sample change. It should be noticed that the mode-locking performance of a fiber laser with net anomalous cavity dispersion is not so dependent on the SA performance if the minimum parameter conditions of the SA used in the laser cavity such as modulation depth and saturation energy are satisfied [81]. We have also checked the long-term stability of the mode-locked fiber laser by observing

the variation of the output pulses for several hours and did not find any substantial variations of the output pulses.

Finally, the output performance of our mode-locked fiber laser was compared to those of mode-locked fiber lasers using SAs based on other types of nonlinear optical materials. The results are summarized in Table 2. Our fabricated ReSe<sub>2</sub>/PVA-based SA exhibited a modulation depth (~3.9%)

**Table 2. Performance Comparison between the Present Work and Previously Demonstrated Mode-Locked Erbium-Doped Fiber Lasers Incorporating Other Saturable Absorption Materials**

Saturable Absorption Materials	SA Threshold Level	Modulation Depth (%)	Wavelength (nm)	3 dB Bandwidth (nm)	Repetition Rate (MHz)	Pulse Width (ps)	Reference
CNT	NA <sup>a</sup>	NA	1556.2	3.7	5.88	0.47	[14]
Graphene	NA	NA	1559	5.24	19.9	0.464	[16]
Graphene	NA	NA	1561.6	1.96	6.99	1.3	[17]
GO	53 W <sup>b</sup>	5.25	1556.5	8.5	17.09	0.615	[19]
Bi <sub>2</sub> Te <sub>3</sub>	44 W	15.7	1547	4.63	15.11	0.6	[22]
Bi <sub>2</sub> Se <sub>3</sub>	12 MW/cm <sup>2c</sup>	3.9	1557.5	4.3	12.5	0.66	[23]
Sb <sub>2</sub> Te <sub>3</sub>	NA	NA	1558.6	1.8	4.75	1.8	[24]
MoS <sub>2</sub>	137 MW/cm <sup>2</sup>	2.7	1556.3	6.1	463	0.935	[26]
WS <sub>2</sub>	600 MW/cm <sup>2</sup>	0.95	1557	2.3	8.86	1.32	[8]
MoSe <sub>2</sub>	24 W	5.4	1555.6	5.4	15.38	0.798	[28]
WSe <sub>2</sub>	NA	0.5	1557.6	2.1	5.31	1.25	[30]
MoTe <sub>2</sub>	NA	1.8	1561	2.4	5.26	1.2	[31]
WTe <sub>2</sub>	64.6 W	2.85	1556.2	4.14	13.98	0.77	[32]
SnS <sub>2</sub>	125 MW/cm <sup>2</sup>	4.6	1562.01	6.09	29.33	0.623	[33]
ReS <sub>2</sub>	74 MW/cm <sup>2</sup>	0.12	1558.6	1.85	5.4801	1.6	[37]
BP	6.55 MW/cm <sup>2</sup>	8.1	1571.45	2.9	5.96	0.946	[46]
Gold nanorod	NA	4.9	1552	3.07	4.762	0.887	[49]
CoSb <sub>3</sub>	8.7 W	5	1557.9	3.44	22.26	0.73	[52]
Ti <sub>3</sub> CN	45 W	1.7	1557	5	15.4	0.66	[53]
ReSe <sub>2</sub>	42 W	3.9	1561.2	3.4	14.97	0.862	This work

<sup>a</sup>NA, not available.

<sup>b</sup>Saturation power.

<sup>c</sup>Saturation intensity.

that is similar to systems based on GO, Bi<sub>2</sub>Se<sub>3</sub>, MoSe<sub>2</sub>, SnS<sub>2</sub>, gold nanorods, and CoSb<sub>3</sub>. In terms of the pulse width of the output pulses, our fiber laser exhibits a reasonable level of the output pulse width (862 fs) for femtosecond laser applications. In terms of saturable absorption properties, ReSe<sub>2</sub> does not exhibit noticeable advantages over other nanomaterials. However, it should be noticed that the ReSe<sub>2</sub> particles used in this work are bulk-structured but not nanostructured. Their sizes were observed to vary from tens of nanometers to around 2 μm. Since bulk-structured materials do not require a complicated fabrication process, we believe that they have significant advantages over nanostructured ones in terms of ease of sample preparation.

## 6. CONCLUSION

We have systematically investigated the nonlinear optical properties of ReSe<sub>2</sub>. The nonlinear absorption coefficient and the nonlinear refractive index of ReSe<sub>2</sub> were measured to be  $-(5.67 \pm 0.35) \times 10^3$  cm/GW and  $-(2.81 \pm 0.13) \times 10^{-2}$  cm<sup>2</sup>/GW, respectively, using the OA and CA Z-scan technique at a wavelength of 1560 nm. The prepared ReSe<sub>2</sub> sample was shown to possess nonlinear saturable absorption and self-defocusing properties. The values of the nonlinear absorption coefficient and the nonlinear refractive index of ReSe<sub>2</sub> are comparable to those of other TMDC materials, as reported in previous studies. A fiberized SA was also fabricated using a side-polished-fiber platform by depositing a ReSe<sub>2</sub>/PVA composite. Using the fabricated ReSe<sub>2</sub>/PVA-based SA, stable mode-locked pulses with a temporal width of ~862 fs could be readily obtained from an EDF laser.

Further investigations for the improvement of the uniformity of the prepared ReSe<sub>2</sub> film are required to enhance the repeatability of the ReSe<sub>2</sub>-based SA fabrication process. We believe that the results of this investigation confirm that ReSe<sub>2</sub> is a good candidate for the development of nonlinear optic or photonic devices.

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