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Nonlinear optical response and applications of tin disulfide in the near- and mid-infrared

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Layered metal dichalcogenides (LMDs) have received considerable attention in optoelectronics and photonics. Tin disulfide (SnS₂) as a member of the LMDs has been employed for transistors, energy storage, and photocatalysts. The optical properties of SnS₂ in the ultraviolet and visible regions have been widely investigated, while the applications of SnS₂ in the near- and mid-infrared regions are still rare. Here, we demonstrate the nonlinear optical response of layered SnS₂ that is exploited as a saturable absorber in the near- and mid-infrared regions. The saturable absorption of SnS₂ is measured at 1.06 and 1.55 µm, which illustrates a low saturable intensity. SnS₂ covered on a D-shaped fiber is used to initiate the mode-locking operations in erbium-, ytterbium-, and thulium-doped fiber lasers and ultrafast pulses are achieved at 1.03, 1.56, and 1.91 µm. These results make SnS₂ an appealing candidate for broadband applications across the near- and mid-infrared regions.

Two-dimensional (2D) materials such as graphene,¹,² black phosphorus (BP),³ and layered metal dichalcogenides (LMDs)⁴⁻⁷ have attracted extensive interest due to their excellent electronic and optical properties.⁸⁻¹⁰ Amongst these 2D materials, graphene, the most well-studied one, has been applied to implement photodetectors,¹¹ modulators,¹² radio-frequency (RF) electronics,¹³ and ultrafast pulse fiber lasers.¹⁴⁻¹⁷ However, the absence of a sizable bandgap in pristine graphene limits its applicability in semiconductor and optoelectronic devices.¹⁸ For example, the graphene-based field effect transistors are difficult to switch off, and engineering a bandgap for graphene would increase fabrication complexity and reduce mobility.¹⁹ In contrast with graphene, BP and LMDs exhibit suitable bandgaps,²⁰⁻²⁵ making them promising alternatives for transistors,²⁰,²¹ photoluminescence,²²,²³ and photocatalysts.²⁴ However, BP would be rapidly degraded in ambient conditions due to its environmental instability.²⁶ Recently, LMDs such as molybdenum disulfide (MoS₂), tungsten disulfide (WS₂), and tin disulfide (SnS₂), have captured much attention due to their diverse electronic, optical, and thermal properties.²⁷⁻³⁰ In recent years, the nonlinear optical characteristics of LMDs have been widely investigated.³¹⁻³⁴ MoS₂ or WS₂-based optoelectronic devices mainly work in the visible region, which is determined by their bandgap.²¹,²³ However, several applications have been demonstrated in the near- (NIR) and mid-infrared (MIR) regions which are beyond their bandgap. For instance, MoS₂ and WS₂ have been utilized as saturable absorbers (SAs) around 1.55 µm.³⁵⁻³⁸

In recent years, SnS₂ has attracted much interest due to its low cost, high chemical stability, excellent photosensitivity, and superior photoelectric properties.³⁹⁻⁴¹ The bandgap of SnS₂ from ~2.033 eV to ~2.4 eV³⁹ is wider than most LMDs, which is crucial in some electronic applications.⁵²,⁴³ The exceptional visible-light-driven photocatalyst based on SnS₂ nanoflakes has been proposed by Umar et al.⁴⁴ Zhong et al. have reported that the SnS₂-based device shows excellent photosensitivity and stability, and its response to the ultraviolet light is very fast.⁴⁵ Additionally, it can be also used in energy storage, photoconduction, and field-emitting devices.⁴¹,⁴⁵ Until now, the study of SnS₂ has mainly focused on its electronic properties and optical applications in the ultraviolet and visible regions.⁴⁶,⁴⁷ However, the optical properties of SnS₂ in the near- and mid-infrared regions are still rarely developed.

In this paper, we demonstrate the nonlinear optical response and applications of layered SnS₂ in the near- and mid-infrared regions. The evanescent field interaction scheme of the propagating light with SnS₂ covered on a D-shaped fiber (SDF) is proposed, not only lengthening the interaction distance between SnS₂ and light but also increasing the optical damage threshold of the SnS₂-covered D-shaped fiber. With the homemade ultrafast fiber lasers, the nonlinear absorption of the SnS₂-covered D-shaped fiber (SDF) in the NIR region is measured. To further confirm the results, SDF is employed as a SA in erbium- and ytterbium-doped fiber (EDF and YDF) lasers to generate ultrafast pulses. In addition, mode-locking operation can also be achieved by the SDF in a thulium-doped fiber (TDF) laser around 2 µm, indicating the saturable absorption of SnS₂ in the mid-infrared (MIR) region.

The schematic of the SDF is shown in Fig. 1(a). The D-shaped fiber is provided with a polished length of 20 mm and the closest distance from the fiber core boundary to the polished surface of 2 µm. The inset of Fig. 1(a) illustrates the structure of monolayer SnS₂ which exhibits a peculiar CdI₂-type crystal structure.⁴⁸ The SnS₂ used in our experiment is...
grown on a sapphire substrate by the chemical vapour deposition (CVD) method. After uniform spin-coating of the SnS2 with PolymethylMethacrylate (PMMA), the large piece of the PMMA/SnS2/sapphire sample is cut into small strips with the length of ~7 mm and the width of ~2 mm. One piece of these small strips is put into a Potassium hydroxide (KOH) solution for several hours to separate the PMMA/SnS2 from the sapphire substrate. Then, the PMMA/SnS2 sheet is transferred into distilled water (DI) in order to remove the residues of the KOH solution. The D-shaped fiber fixed on the glass substrate is immersed in DI, and then we move the D-shaped fiber carefully. When the polished surface of the D-shaped fiber is just under the center of the PMMA/SnS2 sheet, the D-shaped fiber is vertically lifted from the DI and the PMMA/SnS2 covers on the D-shaped fiber. The optical microscopy image shown in Fig. 1(b) displays the transferred PMMA/SnS2 film on the polished surface of the D-shaped optical fiber. The edge of the SnS2 film is marked by an arrow. The SnS2 sample is characterized by Raman spectrometry. As shown in Fig. 1(c), a Raman peak at ~313.8 cm\(^{-1}\) is assigned to the A\(_{1g}\) mode which is similar to those previously reported.\(^{33,47}\) The peak at ~200 cm\(^{-1}\) corresponding to the E\(_{2g}\) mode is undetected, because of the too weak rejection of the Rayleigh scattered radiation to be detected by the Raman sensor.\(^{43}\)

The nonlinear absorption of the SDF sample is investigated as shown in Fig. 2. Figures 2(a) and 2(b) show the nonlinear absorption of SDF, which are experimentally implemented by homemade ultrafast fiber lasers (pulse duration of ~2 ps) with center wavelengths of ~1.06 and ~1.55 \(\mu\)m, respectively. The solid curves are the fitting results of experimental data on the basis of a simplified two-level saturable absorption model.\(^{49}\) As illustrated in Figs. 2(a) and 2(b), SDF exhibits the typical characteristics of saturable absorption that the absorption decreases with increasing the pulse intensity. At 1.06 \(\mu\)m, the saturable absorption \(\alpha_0\), the non-saturable absorption \(\alpha_{\text{nonsat}}\), and the saturation intensity \(I_{\text{sat}}\) are 0.6%, 58.3%, and 10.9 MW/cm\(^2\). At 1.55 \(\mu\)m as shown in Fig. 2(b), \(\alpha_0\), \(\alpha_{\text{nonsat}}\), and \(I_{\text{sat}}\) are 5%, 44.4%, and 19.2 MW/cm\(^2\), respectively. Our experimental results indicate that SnS2 can be used as a broadband SA in the NIR region, where the photon energy is lower than the bandgap of the perfect monolayer or bulk SnS2. The saturable absorption in the NIR region should be attributed to the sub-bandgap absorption which originates from the atomic defect in SnS2.\(^{50,51}\)

As the saturable absorption of SDF has been demonstrated in the NIR region, SDF is here applied as a SA in YDF and EDF lasers. Two fiber lasers have the same ring cavity configuration that consists of a pump source, a wavelength-division multiplexer (WDM), a polarization independent isolator (PI-ISO), an optical coupler (OC) with 10% output ratio, a polarization controller (PC), and a SDF SA, which can be seen in Fig. 3. YDF (~0.6 m) and EDF (~7 m) with dispersion parameters of ~33 ps/nm/km and ~9 ps/nm/km are pumped by a 976 nm laser diode. In the YDF laser, the total length and the net dispersion of the cavity are given as ~54 m and ~1.19 ps\(^2\), respectively. Under the normal dispersion condition, the fiber laser tends to emit dissipative solitons (DSs) and the balance between gain and loss plays an important role in DS evolution dynamics.\(^{52}\) In the EDF laser, the total cavity length is ~47 m and the net dispersion is ~0.778 ps\(^2\). Under the anomalous dispersion condition, conventional soliton (CS) can be achieved due to the balance between dispersion and nonlinearity.\(^{53,54}\)

For the EDF laser, by increasing the pump power to ~45 mW, self-started mode-locking operation is achieved when the polarization controller is adjusted appropriately. At this time, multiple pulses circulate in the laser cavity. The bound-state and hysteresis phenomenon can be observed, similar to the reports in Refs. 55 and 56. When the pump power is decreased from ~45 mW to ~9 mW, the single-pulse mode-locking state can be obtained. Figure 4(a) demonstrates a typical spectrum of CS, with a central wavelength of ~1561 nm. Several sidebands are distributed on both sides of the spectrum, which is the typical characteristic of a soliton pulse.\(^{57}\) Figure 4(b) illustrates the autocorrelation trace (AC), which has a full width at half maximum of 2.51 ps. Assuming a sech\(^2\) temporal profile, the pulse duration is estimated as 1.63 ps. The time bandwidth product (TBP) is calculated as 0.321, indicating that the pulse is slightly chirped.
The pulse train depicted in Fig. 4(c) shows that the separation between adjacent pulses is \( \Delta t = 227.3 \text{ ns} \). The radio-frequency (RF) spectrum is shown in Fig. 4(d). The fundamental repetition rate is 4.397604 MHz and the peak-to-background ratio is above 60 dB.

When SDF is employed in the YDF laser, DSs can be emitted with appropriate pump power and polarization state, as shown in Fig. 5. The optical spectrum of DSs in Fig. 5(a) is centered at \( \lambda = 1031 \text{ nm} \) with a 3-dB spectral width of \( 1.2 \text{ nm} \), which displays the similar spectral profile to that in Refs. 58 and 59. Figure 5(b) plots a typical second harmonic generation AC, which is well fitted by a Gaussian function. The pulse duration is calculated as \( \sim 282 \text{ ps} \). The corresponding TBP is calculated as 95.5, which means that the pulse is highly chirped. The pulse train and fundamental repetition rate are given in Figs. 5(c) and 5(d).

The applications of SnS\(_2\) are further investigated in the MIR region. The same ring cavity configuration in Fig. 3 is used to construct a TDF laser working around 2 \( \mu \text{m} \). The TDF is pumped by an EDF laser emitting at 1.57 \( \mu \text{m} \). The total length of the cavity is \( L = 1 \text{ m} \) and the net cavity dispersion is about \( -6.22 \text{ ps}^2 \). The mode-locking operation is obtained with the proper pump power by adjusting PC. As shown in Fig. 6(a), the mode-locked optical spectrum has a center wavelength of \( \sim 1910 \text{ nm} \) with a 3-dB spectral width of \( \sim 1.2 \text{ nm} \). Figure 6(b) shows that the repetition rate is 1.987646 MHz, corresponding to 502.5 ns of round-trip time (inset of Fig. 6(b)). The peak-to-background ratio of the RF spectrum is above 60 dB, indicating a low-amplitude fluctuation and good mode-locking stability. The experimental results indicate that SnS\(_2\) exhibits saturable absorption in the MIR region, and the SDF can work as a good mode locker to achieve mode-locking operation around 2 \( \mu \text{m} \).

In conclusion, we explore the nonlinear optical properties of SnS\(_2\) in the near- and mid-infrared regions. The saturable absorption of SnS\(_2\) is measured with ultrafast fiber lasers at 1.06 and 1.55 \( \mu \text{m} \). Based on SDF SA, mode-locked fiber lasers are achieved at 1.03, 1.56, and 1.91 \( \mu \text{m} \) with the single pulse energy of about 492, 33.2, and 513 pJ, respectively. Compared with fiber lasers based on other LMD materials, the SnS\(_2\)-based fiber lasers show a rather small mode-locking threshold derived from the low saturable absorbing threshold of the SDF SA. The laser performance can be further optimized by improving the fabrication process of SDF SA. The Q-switching operation cannot be observed in the experiment for all three different fiber lasers. It should be attributed to the relatively small modulation depth of the SDF SA. It is worth mentioning that the SDF SA is not damaged even at the maximum incident pump power, which further confirms the high damage threshold of SDF SA.

In order to testify the stability of the mode-locked fiber lasers, we observe the fiber lasers while they are operating continuously for several hours and the spectra show no obvious changes. According to experimental results, the performance of the proposed SA is comparable to those based on other 2D materials. Our works demonstrate the applications of SnS\(_2\) beyond its bandgap, which paves the way for SnS\(_2\)-based opto-electronic devices in a broadband wavelength.

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