

Characteristics of saturable absorption of MoS₂ films in the visible to near-infrared range

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Abstract: MoS₂ films consisting mainly of few-layer MoS₂ nanoplatelets are fabricated. Tunable Z-scan measurement reveals strong saturable absorption (~70% modulation depth) around 500 nm, demonstrating the potential of MoS₂ for visible laser mode-locking and optical switching.

OCIS codes: (320.7080) Ultrafast devices; (320.7090) Ultrafast lasers; (140.4050) Mode-locked lasers

1. Introduction

Saturable Absorbers (SAs) are key enabling elements for mode-locked lasers which are finding ever wide-ranging applications in fields such as optical communication, spectroscopy, biomedical diagnosis and precision material processing. Key requirements for SAs include wide operation wavelength range, large nonlinear response (or modulation depth), ultrafast carrier relaxation time, compactness as well as the ease of tailoring the relevant optical parameters. Carbon nanomaterials including carbon nanotubes and graphene have proved to be excellent saturable absorbers for fiber and solid-state lasers [1-5]. They have become important alternative to the more established SESAM technology, especially at longer wavelength range, i.e. mid infrared. Although both graphene and carbon nanotubes exhibit broadband saturable absorption, their applications in ultrafast photonics have so far been confined in the near-infrared to mid-infrared range [3-7]. Employing the E₂₂ transition in nanotubes is a feasible way to extend carbon nanotubes' nonlinear absorption edge towards shorter wavelength, though it leads to higher saturation intensity and no characterization of nonlinear absorption in the visible range has been reported so far [8].

Molybdenum disulfide (MoS₂) is subject to intense research efforts because of its unusual electronic and optical properties. While few-layer and bulk MoS₂ is a semiconductor with an indirect bandgap of ~1.3 eV, monolayer MoS₂ has a direct bandgap of ~1.8 eV [9]. Recently, saturable absorption has been reported for MoS₂ dispersions [10,11], and mode-locked lasers at ~1 μm has also been reported [11,12], suggesting a broad nonlinear absorption bandwidth. However, thus far there has been no report that systematically studies the wavelength dependence of the wideband nonlinear absorption of MoS₂, especially for a solid-state form of MoS₂. In this paper, we have fabricated MoS₂ films using a vacuum filtration technique. The film can be easily transferred to other optical surfaces, such as an optical fiber ferrule, and its linear and nonlinear absorptions can be readily tuned by controlling the film thickness. Investigation in the nonlinear optical properties by Z-scan measurement employing ~100 fs pulses tunable across visible to near infrared is carried out. Our results show that few-layer MoS₂ nanoplatelets exhibit strong saturable absorption (~70% modulation depth) in the visible range and the modulation depth (in transmittance) diminishes with the increase of exciting wavelength. Our results show that MoS₂ films may be an excellent SA material for ultrafast pulse generation and optical switching in the visible range where no competent SA technology exists yet.

2. MoS₂ film fabrication

Two-dimensional (2D) MoS₂ dispersions were prepared by liquid exfoliation using ionic surfactant sodium cholate as a stabilizer [13]. First of all, 200 mg of MoS₂ powders (Sigma-Aldrich) were dispersed in 40 mL of 1.5 mg/mL sodium cholate (SC) (Sigma-Aldrich) aqueous solution, followed by sonication for 1 hour with high intensity ultrasonic liquid processor. Then, the obtained MoS₂ suspensions were centrifuged at 3000 r/min for 90 minutes. The top 3/4 of the dispersion was collected by pipette for later use. 2 mL of the above prepared 2D MoS₂ dispersion was taken and diluted into 50 mL with a bath sonication for 1 hour. The diluted MoS₂ dispersion was vacuum-filtrated using a mixed cellulose ester membrane with 200 nm pores (Millipore). The film was then transferred onto a quartz substrate and heated at 60 °C for 6 hours. To remove the cellulose ester membrane, the MoS₂ films were immersed in acetone for several times. Fig. 1(a) shows the absorption spectrum of the MoS₂ film. Four typical peaks can be observed in the range of 300 ~ 800 nm, which correspond to the transition of excitons of A, B, C, D, respectively.

Fig.1(b) shows the Raman spectrum of the MoS₂ film. While the mean difference for E_{2g}¹ and A_{1g}¹ mode is similar to the bulk MoS₂ value of ~ 25.2 cm⁻¹, the broadening of the linewidths of the E_{2g}¹ band (2.9 cm⁻¹) and the A_{1g} band

(3.5 cm^{-1}) in comparison with those of the bulk MoS_2 (i.e. $\sim 1.4 \text{ cm}^{-1}$ and $\sim 3.2 \text{ cm}^{-1}$ respectively) verifies the significant reduction of the flake thickness from the bulk MoS_2 [10]. Fig.1(c) shows a photograph of an as-fabricated MoS_2 film (on a quartz substrate). The typical thickness of the film is 100-300 nm.

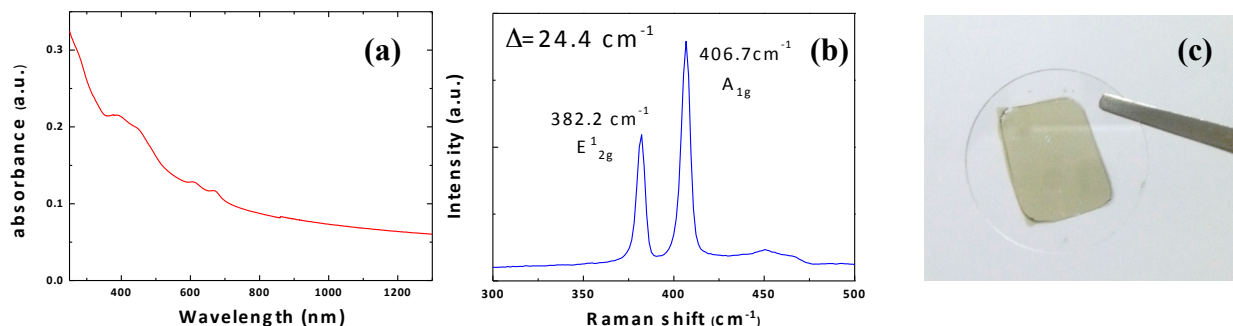


Fig.1 (a) UV-Vis-IR absorbance curve; (b) Raman spectrum of the MoS_2 film; (c) Photograph of a MoS_2 sample.

3. Wavelength tunable Z-scan characterizations

To gain insights into the broadband nonlinear optical properties of MoS_2 , we employ an open-aperture Z-scan setup to characterize the nonlinear absorption of MoS_2 nanoplatelets. The pump source consists of a Coherent Libra-HE amplifier system with a OPERA-SOLO Optical parametric amplifier (OPA). Specifically, we have chosen the wavelength 500 nm, 650 nm, 800 nm, 950 nm and 1100 nm to illustrate the trends of the wideband nonlinear absorption. The excitation pulse duration is $\sim 100 \text{ fs}$ (with a 1 kHz repetition rate) and a neutral density filter is employed to set the incident optical power. A power meter with a pair of photodiode detectors (Ophir PD300-IR) is used to record the incident and transmitted power through the samples. At each excitation wavelength, we have attempted to investigate the full range of the available pump power from the OPA. However, to avoid laser damage caused by either high peak intensity or thermal effects, we have limited our excitation power to a range within which Z-scan curves are readily reproducible without change in linear transmittance of the samples [11]. It should be noticed that two-photon absorption (TPA) threshold is found to be higher than the stability range of excitation power, that is why we concern ourselves only with saturable absorption in this experiment.

Fig.2 illustrates the Z-scan results for a set of excitation wavelengths from 500 nm - 1100 nm (with a 150 nm interval). The Z-scan curves under different excitation intensities indicate a large nonlinear response up to a modulation depth of $\sim 70\%$ at 500nm. With the increase of the excitation wavelength, we observe continuous decrease of the modulation depth.

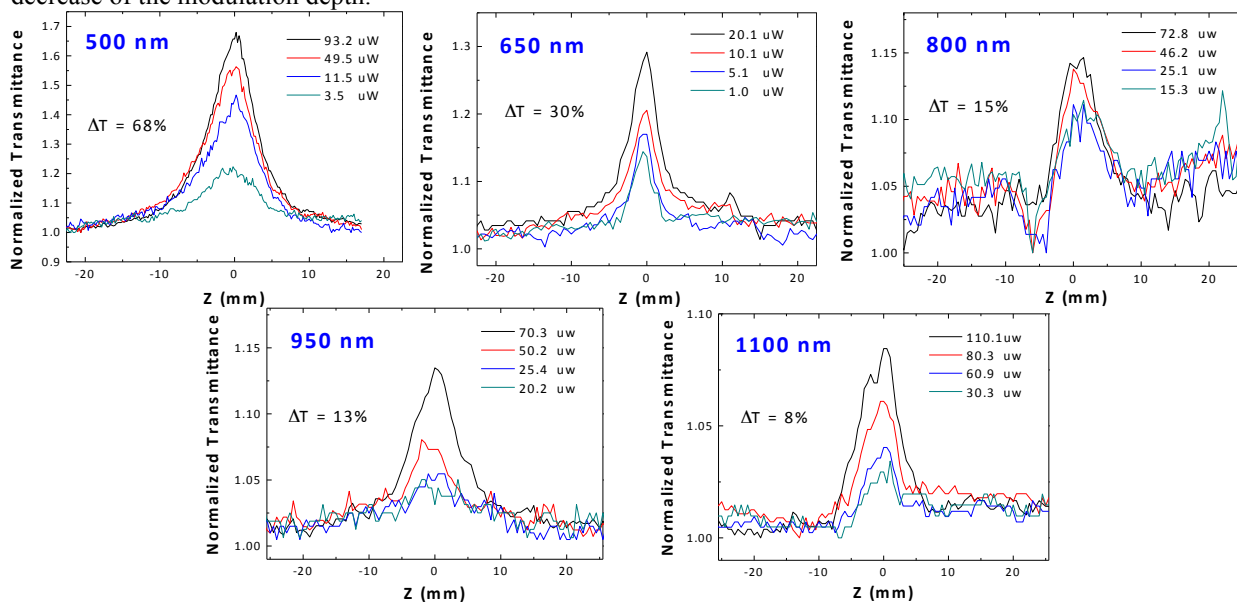


Fig.2 Normalized transmittance at different excitation wavelengths (500nm, 650nm, 800nm, 950nm, 1100nm).

4. Conclusion

We have for the first time systematically investigated the characteristics of wideband saturable absorption of MoS₂ nanoplatelets (in the form of a thin film) in the visible and near-infrared range. Using an open-aperture Z-scan technique, the MoS₂ sample is found to exhibit strong and wideband nonlinear optical properties, with the best signals obtained at a wavelength ~ 500 nm. The modulation depth at ~500 nm are measured to be ~ 70%. Our results show that MoS₂ has great potential in visible laser mode-locking/Q-switching and optical switching applications.

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