Vertically standing PtSe2 film: a saturable absorber for a passively mode-locked Nd:LuVO4 laser

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The novel vertically standing PtSe2 film on transparent quartz was prepared by selenization of platinum film deposited by the magnetron sputtering method, and an Nd:LuVO4 passively mode-locked solid-state laser was realized by using the fabricated PtSe2 film as a saturable absorber. The X-ray diffraction pattern and Raman spectrum of the film indicate its good crystallinity with a layered structure. The thickness of PtSe2 film is measured to be 24 nm according to the cross-section height profile of the atomic force microscope image. High-resolution transmission electron microscopy images clearly demonstrate its vertically standing structure with an interlayer distance of 0.54 nm along the c-axis direction. The modulation depth (ΔT) and saturation fluence (Φs) of PtSe2 film are measured to be 12.6% and 17.1 μJ/cm2, respectively. The obtained mode-locked laser spectrum has a central wavelength of 1066.573 nm, with a 3 dB bandwidth of 0.106 nm. The transform limited pulse width of the mode-locked laser was calculated to be 15.8 ps. A maximum average output power of 180 mW with a working repetition rate of 61.3 MHz is obtained. To the best of our knowledge, this is the first report of the generation of ultrafast mode-locked laser pulses by using layered PtSe2 as a saturable absorber.

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1. INTRODUCTION

Recently, two-dimensional (2D) transition metal dichalcogenides (TMDs) have attracted tremendous attention due to their distinctive properties, e.g., tunable bandgap, high carrier mobility, and strong nonlinear optical properties [1–3], which may bring revolutionary changes in diverse fields such as electronics, photonics, catalysis, laser, renewable energy, and so on [4–8]. As a newly developed group-10-based TMD, PtSe2 has many attractive properties for device applications. Monolayer PtSe2 is a semiconductor with a bandgap of 1.2 eV, and it becomes smaller by increasing the layer number; the bulk PtSe2 turns to be semimetal with a zero bandgap [9]. The carrier mobility of PtSe2 has been predicted to be the highest among the TMD materials, being comparable to that of black phosphorus (BP) [10]. In addition, PtSe2 has excellent stability in the air [9,11]. Because of these appealing properties, the research work related to 2D PtSe2 has attracted great attention recently, and it has been applied and studied in field-effect transistors [12], photodetectors [13,14], photocatalysis, and so on [15]. However, so far, to the best of our knowledge, no research work has been reported on its application in mode-locked lasers for ultrafast laser generation.

Passively mode-locked lasers are considered among the high-end laser photonic products, as they can produce ultrafast lasers with high peak power. These laser systems have many important applications, such as laser surgery, ultrafine laser micromachining, high-accuracy measurement, and ultrafast pump sources for scientific research. It is well known that a saturable absorber (SA) is the critical component of the passively mode-locked laser system, which can convert a laser operating in continuous wave mode into pulse mode by simply inserting a saturable absorber within the laser cavity. So far, many different materials have been developed as saturable absorbers for laser mode locking, including semiconductor saturable absorber mirrors (SESAMs) [16], carbon nanotubes (CNTs) [17], graphene [18–21], TMDs [22,23], topological insulators [24,25],...
BP [26–30], Mxene [31], and so on. Among them, SESAM has been in commercial use, but it also has some disadvantages, such as complicated fabrication processes, high cost, and narrow operating bandwidth. The saturable absorbing performance of CNTs depends too much on its structure, such as number of wall layers, diameter, and length of CNT. Graphene, a single layer of carbon atoms, was used as a potential saturable absorber for generating ultrashort pulsed laser due to its ultrafast exciton recovery time [32]. However, graphene has a small modulation depth because of the weak layer absorption. TMDs and BP are newly developed saturable absorber materials. They have advantages such as tunable bandgap, high carrier mobility, and high modulation depth. However, they have some drawbacks; for example, BP is very unstable in air due to oxidation; TMDs, such as MoS2 and WS2, have long exciton recovery time, making them difficult to obtain ultrashort pulses. Therefore, developing novel 2D materials for saturable absorbers is still a very active and important research field. Additionally, our research findings will also benefit research work in other related fields, e.g., nonlinear optics and microphotonics.

In this work, wafer-scale vertically standing polycrystalline PtSe2 film was prepared through the magnetron sputtering deposition method using transparent quartz as the substrate. The prepared PtSe2 film was applied in the solid-state mode-locked Nd:LuVO4 laser system. The obtained mode-locked laser spectrum has a central wavelength of 1066.573 nm, with a 3 dB bandwidth of 0.106 nm. A maximum average output power of 180 mW with a working repetition rate of 61.3 MHz was obtained at an incident pump power of 3.64 W.

2. EXPERIMENTAL SECTION

A. Materials Synthesis and Characterization

PtSe2 films on quartz substrates in this work were grown by a simple selenization method. In brief, platinum films were first deposited on SiO2/Si (300 nm SiO2 thickness) using the magnetron sputtering system. The metal-deposited SiO2/Si substrates were then placed at the center zone of the furnace, and selenium powder (99.99% purity) was placed at the upstream side. Selenium was evaporated at 220°C dragged by 50 SCCM (standard cubic centimeter per minute) argon flow. The central temperature of the tube furnace was set to 420°C. After selenization for an hour, gray films form on the substrates. The Raman spectrum measurements were carried out on a HORIBA Raman spectrometer with a 488 nm argon ion laser. The X-ray diffraction (XRD) pattern was recorded using a Rigaku SmartLab X-ray diffractometer. The topography of samples was obtained by atomic force microscopy (AFM, VeecoNanoscope V). The morphology, crystal structure, and chemical composition were investigated using a field emission transmission electron microscope (FETEM, JEOL Model JEM-2100F), equipped with an energy-dispersive spectrometer (EDS).

B. Laser Mode-Locked Characterization

The experimental setup of the passively mode-locked laser using PtSe2 as a saturable absorber is shown in Fig. 1. A folded cavity was used in our experiment. The pump source is an 808 nm fiber-coupled diode laser with a maximum power of 35 W, a core diameter of 105 μm, and a numerical aperture of 0.22. Nd:LuVO4 crystal has been widely used as a laser medium for diode-pumped solid-state lasers because of its high doping concentration and large absorption cross section. The Nd:LuVO4 crystal has an antireflection (AR) coating of around 1.06 μm and 808 nm. It has an Nd3+ concentration of 0.5% and dimensions of 3 mm × 3 mm × 8 mm. The M1, M2, M4, and M6 are flat mirrors, while M3 and M5 are concave mirrors with curvature radii of 1000 and 300 mm, respectively. M2 is AR coated at 808 nm (T > 95%) and high-reflection (HR)-coated at around 1.06 μm (R > 99.8%). The output coupler M1 has a transmission of 3% at 1.06 μm. The other mirrors are HR-coated at around 1.06 μm (R > 99.8%). The length of the cavity is about 2.45 m. The distance between the mirror M6 and the PtSe2 SA is about 3 mm, as illustrated in Fig. 1.

3. RESULTS AND DISCUSSION

The XRD pattern of the PtSe2 film is presented in Fig. 2(a), showing a strong diffraction peak located at 16.82°, which corresponds to the (001) planes arranged in the direction perpendicular to the c axis of PtSe2, indicating layer structure of the as-prepared PtSe2. Besides, three more weak diffraction peaks located at 27.76°, 32.76°, and 49.24° are detected and shown in the inset of Fig. 2(a), which are indexed to (100), (101), and (110) planes of PtSe2, respectively [33]. Figure 2(b) shows the Raman spectrum of PtSe2 film. Three prominent peaks are identified at 174 cm−1, 204 cm−1, and 228 cm−1, corresponding to E2g, A1g, and A2g vibration modes, respectively. The E2g mode is an in-plane vibration mode of platinum and selenium atoms, while the A1g and A2g modes are out-plane vibration modes of selenium atoms [11]. These strong characteristic Raman peaks indicate that the prepared PtSe2 film has good crystallinity, agreeing well with the XRD result. Figure 2(b) inset is a Raman mapping image of the E2g peak intensity of PtSe2 film obtained within an area of 10 μm × 10 μm. It shows the continuous and homogeneous signal of E2g, indicating the good continuity and homogeneity of PtSe2 film. An atomic force microscopy image of PtSe2 film is presented in Fig. 2(c), showing the clear border of the film. The cross-section height profile given in Fig. 2(d) shows the thickness of the film is about 24 nm.
Figure 3(a) presents a photograph of the PtSe$_2$ film grown on a quartz substrate with a size of 1.5 cm × 1.5 cm, showing it is a semitransparent gray-colored film. The transmission electron microscope (TEM) images of the PtSe$_2$ film with different magnifications are shown in Figs. 3(b) and 3(c), revealing that the synthesized PtSe$_2$ is continuous and dense polycrystalline with a vertically standing layered structure. Its high-resolution transmission electron microscope (HRTEM) image presented in Fig. 3(d) demonstrates the layer structure, with an interlayer distance of 0.54 nm, agreeing well with the interlayer distance of (001) planes, and confirming the vertically grown mode of PtSe$_2$ on the quartz substrate. Figure 3(e) is the selected area electron diffraction (SAED) pattern of PtSe$_2$ film, showing four clear diffraction rings, from inside to outside, which can be readily ascribed to the (001), (100), (101), and (110) planes of PtSe$_2$, respectively. The detected four groups of planes agree well with the above XRD results. Figure 3(f) shows the EDS spectrum of the film, demonstrating the film is composed of two elements, platinum and selenium, with no impurity.

A pulsed laser with a pulse width of 25 ps working at 1.06 μm was used to investigate the nonlinear optical properties of the PtSe$_2$ film. The relationship between the transmission and the input pulse fluence is shown in Fig. 4(a).
The modulation depth (ΔT) and saturation fluence (Φs) are determined to be 12.6% and 17.1 μJ/cm², respectively. The as-prepared PtSe₂ film was used as a saturable absorber in an Nd:LuVO₄ laser system. The mode-locked pulse trains were recorded by a fast photodiode connected to a digital oscilloscope (Agilent DSO7104B) and are shown in different time scales in Figs. 4(b)–4(d). The pulse trains recorded on the oscilloscope show uniform intensity. The measured repetition rate is 61.3 MHz, with a pulse-to-pulse interval of 16.3 ns, corresponding to the cavity round-trip time, indicating the laser is under mode-locking operation. Much work has been done by using Group-6 TMDs, including MoS₂, WS₂, MoSe₂, WSe₂, etc. as saturable absorbers for generating pulsed lasers [34,35]. However, most of the laser mode-locking demonstrations are based on fiber lasers, while for solid-state lasers, the realization of mode-locked operation based on TMDs is rarely reported. However, as is well known, a crystal laser can be a better gain medium for high-pulse energy generation due to its large mode volume for energy storage. Currently, Q-switched microsecond or nanosecond lasers are usually obtained in solid-state lasers based on TMDs [36–39]. In this work, it is exciting to report the realization of solid-state mode-locked laser using PtSe₂ as the saturable absorber.

Figures 5(a) and 5(b) show the output laser spectra measured before and after mode locking, respectively, using an optical spectrum analyzer with a wavelength resolution of 0.01 nm. Their central wavelengths are 1066.572 and 1066.573 nm, respectively, nearly the same. The 3 dB bandwidth of the spectrum before mode locking is 0.035 nm, while it is 0.106 nm for the one after mode locking. The transform limited pulse width of the mode-locked laser was calculated to be 15.8 ps using the formula

\[ t_p = \frac{0.441 \lambda^2}{\Delta \lambda C}, \]

where \( t_p \) denotes the pulse width of the mode-locked laser, \( \Delta \lambda \) represents the wavelength bandwidth of the mode-locked laser, \( \lambda \) is the central wavelength of the laser, \( C \) is the speed of light, and 0.441 is the transform limited time-bandwidth value of a Gaussian shape pulse. Figure 5(c) is the curve of the average output power with respect to the incident pump power. The stable mode-locked operation was observed at an incident pump power of 2.23 W. The maximum average output power of the mode-locked laser can reach 180 mW at an incident pump power of 3.64 W. The calculated pulse energy is 2.94 nJ.

4. CONCLUSIONS

In summary, we believe this is the first report of the successful generation of a stable mode-locked pulsed laser by using the novel layered PtSe₂ as a saturable absorber. The XRD pattern and Raman spectrum indicate the layered structure and good crystallinity of PtSe₂ film prepared by the magnetron sputtering method. The HRTEM image clearly presents its vertically grown mode on the quartz substrate. The modulation depth (ΔT) and saturation fluence (Φs) of PtSe₂ film are measured to be 12.6% and 17.1 μJ/cm², respectively. Stable and uniform pulse trains are obtained from a PtSe₂-based mode-locked laser, and the 3 dB bandwidth of the mode-locked laser output spectrum is measured to be 0.106 nm. The transform limited pulse width of the mode-locked laser is calculated to be 15.8 ps. The maximum average output power of the mode-locked laser can reach 180 mW, and the working repetition rate is 61.3 MHz. PtSe₂ has been explored as a completely new saturable absorber for generating mode-locked lasers in this research.

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