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Watt-level high-stability all-solid-state passively Q-switched laser based on germanene nanosheets

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Excellent Q-switching operations modulated by new two-dimensional (2D) saturable absorber (SA) materials with stable performance is a hot topic in all-solid-state pulsed laser research. In this work, the watt-level high-stability passive Q-switching operation in a solid-state Nd:YVO_4 laser utilizing the 2D germanene nanosheets as SA was first realized. The nonlinear optical properties of the germanene nanosheets (Ge-Ns) were characterized by experimental means. The stable Q-switched pulse sequence was acquired with a 60.6 ns narrowest pulse width and a 528.6 kHz maximal repetition rate. The average output power of 0.965 W and the corresponding pulse peak power of 30.12 W are obtained under the pump power of 7 W. The findings of the experiments demonstrate that germanene material has remarkable nonlinear optical properties and can be used as an excellent saturable absorber in the field of optical pulse modulation.

KEYWORDS

Germanene, passively Q-switched, solid-state laser, high peak power, high stablity

Introduction

Short pulse solid-state Q-switched lasers have a very wide range of applications in military, material processing, clinical applications, and nonlinear optics due to their high pulse energy and simple manufacturing [1–3]. At present, the Q-switching technique can be divided into passive Q-switched and active Q-switched according to the control mode. By contrast, the passive Q-switched technology has the advantages of simple structure and low cost [4, 5]. However, the saturable absorber (SA) is one of the key devices of passively Q-switched lasers. Extensive research has been carried out to better SAs since the first successful commercialization of semiconductor saturable absorber mirrors (SESAMs) in 1992 [6]. So far, in addition to SESAMs, many new two-dimensional (2D) nanomaterials have also been gradually proved to have excellent saturable absorption characteristics, such as graphene [7, 8], carbon nanotubes [9–11], topological insulators (TIs) [12, 13], transition metal dichalcogenides (TMDs) [14–17], MXenes [18, 19], Xenes [20–22], and black phosphorus (BP) [23–25]. Despite the fact that these 2D materials have been

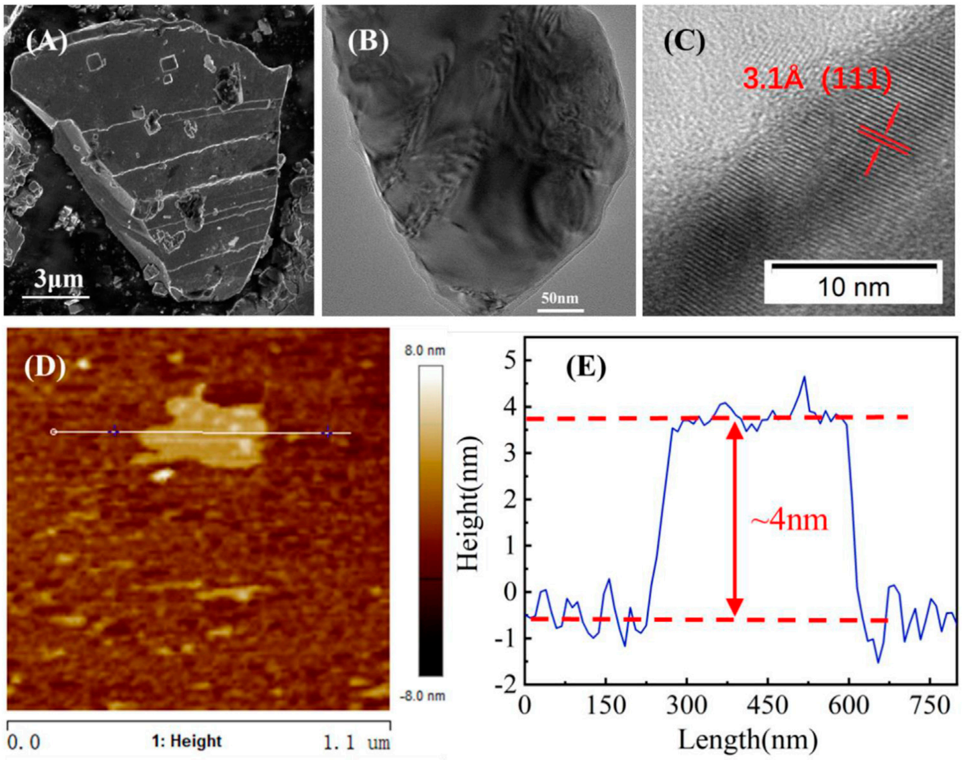

FIGURE 1
Characterizations of few-layer Ge-Ns. **(A)** SEM image of the germanene crystal powder sample; **(B,C)** HRTEM image with higher resolution; **(D)** AFM images; and **(E)** corresponding thickness profile of Ge-Ns.

employed effectively as broadband SAs, there are still some drawbacks that limit their applications in all-solid-state lasers. For example, the smaller the modulation depth (MD) (~2.3% for monolayer) of graphene, the larger the bandgap of TMDs, and the poorer the stability of BP [26–28]. Therefore, the exploration of new SA materials with better performance is necessary for Q-switched or mode-locking lasers.

As a germanium-based analog of graphene, germanene has been theoretically and experimentally proven to have the advantages of environmental friendliness, facile fabrication, and broadband absorption, indicating promising characteristics for optoelectronic fields [29]. In 2009, Cahangirov et al. first demonstrated that germanium has a stable honeycomb structure [30]. In 2014, Li et al. experimentally synthesized 2D germanene sheets on the Pt surface by electron beam evaporation technology [31]. In the same year, atom-thin, ordered 2D multiphase germanium film was prepared by Davila et al. using molecular beam epitaxy technology on the surface of gold [32]. In 2022, Sun et al. for the first time used germanene nanoplates as SA to realize the harmonic mode-locking operation with the order of fundamental frequency, second, fourth, fifth, and sixth based on the germanene–PVA film in an erbium-doped fiber laser [33]. Very recently, by the liquid-phase exfoliated method, the germanene nanosheets are prepared and used for the generation of ultrashort pulses in fiber lasers at 1,061.1, 1,559.3, and 1883.5 nm, respectively [34]. As a natural quantum well structure, the semiconductor layer of germanene acts as a reservoir, and the carriers in the layer have a substantially shorter relaxation period [29]. The optical bandgap of the germanium nanosheets has been calculated to be about 2.46 eV, corresponding to the wavelength of ~506 nm [34]. The broadband saturable absorption of the germanene nanosheets indicates the existence of sub-bandgap absorption, which may be caused by the high edge to surface area ratio of 2D germanene nanosheets [35]. Moreover, atomic vacancy defects can also reduce the bandgap and hence make contributions to the sub-bandgap absorption [36]. The nonlinear absorption coefficient of germanene nanosheets is much higher than that of graphene and has superior environmental durability to BP [37]. All the studies so far show that germanene is a kind of optical material with excellent properties. However, the research on the pulse modulation characteristics of germanene in solid-state lasers is still insufficient, and the relevant research on germanene in solid-state Q-switched lasers has not been found yet.

In this work, using self-made germanene nanosheets (Ge-Ns) as SA, a passively Q-switched $Nd:YVO_4$ laser with high stability

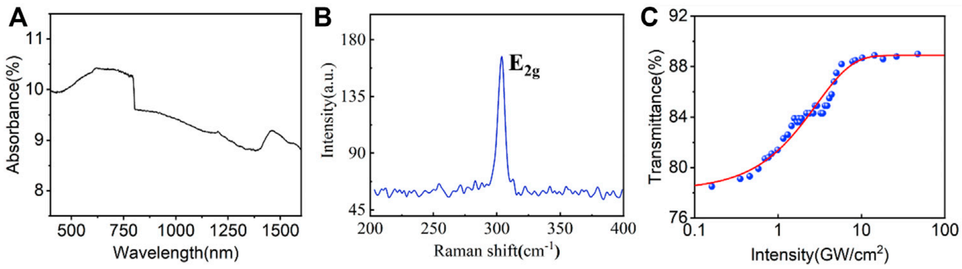

FIGURE 2
(A) Absorption spectrum; **(B)** Raman spectrum; and **(C)** nonlinear optical response of the Ge-N SA.

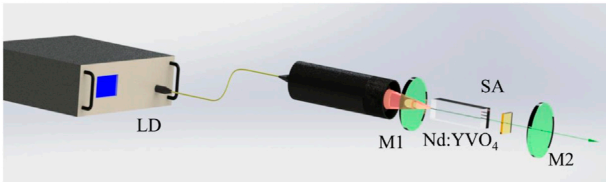

FIGURE 3
Schematic setup of the Nd:YVO_4 laser with germanene as the saturable absorber. LD: laser diode; SA: Ge-N saturable absorber.

was realized for the first time. The nonlinear optical parameters of the germanene nanosheets are characterized. Stable Q-switched pulses were obtained with the narrowest pulse width of 60.6 ns and the corresponding repetition rate of 528.6 kHz at the pump power of 7 W. The single pulse energy and peak power of 1.83 μJ and 30.12 W are calculated, respectively. As far as we know, this is the first time that the saturable absorption feature of germanene SA in a solid-state laser was demonstrated, indicating its capability of generating high stable Q-switched pulses in an all-solid-state laser.

Preparation and characterization of germanene saturable absorber

Using the liquid-phase exfoliated (LPE) method, the preparation process of 2D Ge-Ns is similar to our previous study [15]. The 2D germanene dispersion is ultrasonic for 10 h and centrifuged for 15 min at 5,000 rpm to remove the large size germanium crystals. Then, the desired 2D Ge-N solution was obtained. The rotary coating evaporation technology with a spin coating speed of 300 rpm was used to deposit Ge-Ns on the facet of an uncoated sapphire substrate.

The microscopic morphology and saturable absorption properties of the Ge-Ns are characterized. Figure 1A shows the configuration of the surface of the Ge crystal powder sample as seen through a field-emission scanning electron microscope (SEM, Sigma 500, Zeiss). The picture shows solid agglomerates of several microns with distinct layered structures which include the cleavage plane and cleavage step, indicating that the bonding force between layers is a weak van der Waals force. Figure 1B shows a picture of the prepared Ge-Ns measured using a high-resolution transmission electron microscope (HRTEM, JEM-2100). It can be seen that Ge-Ns are very thin and almost transparent. At a resolution of 50 nm, the layered structure of Ge-Ns can be easily observed, which is very different from Figure 1A. Changing the HRTEM resolution to 10 nm, the lattice spacing of 3.1 Å of the few-layer Ge-Ns can be observed from the HRTEM image, as shown in Figure 1C. The atomic force microscope (AFM, Bruker Multimode 8) was used to measure the thickness of the Ge-N sample. Figure 1D exhibits the obtained AFM image. The corresponding thickness profile of the sample is given in Figure 1E. It can be found that the average thickness of the sample is about ~ 4 nm, corresponding to 13 layers [38].

Figures 2A,B show the UV-Vis-NIR absorption spectrum and Raman spectrum of the Ge-N sample. The broadband absorption characteristic curve is measured from 400 to 1600 nm using a UV-Vis-NIR spectrophotometer. At 1064 nm, the light absorption rate of the Ge-N sample is 9.1%. The Raman spectrum was measured to analyze the chemical structure and vibration mode of the Ge-N sample. From the diagram, the prominent peak (E_{2g}) at 304.4 cm^{-1} can be observed, which is consistent with the previous reports [29, 34].

The nonlinear transmittance of Ge-N SA is measured by the double-optical-path method [15]. A solid-state Q-switched laser at 1.06 μm (120 ns pulse width, 15 kHz 95 repetition rate) was used as the laser source for testing. The experimental data of transmission *T(I)* shown in Figure 2C with symbols are fitted by the formula as follows:

$$T(I) = 1 - T_{ns} - \Delta T * exp\left(-\frac{I}{I_{sat}}\right).$$

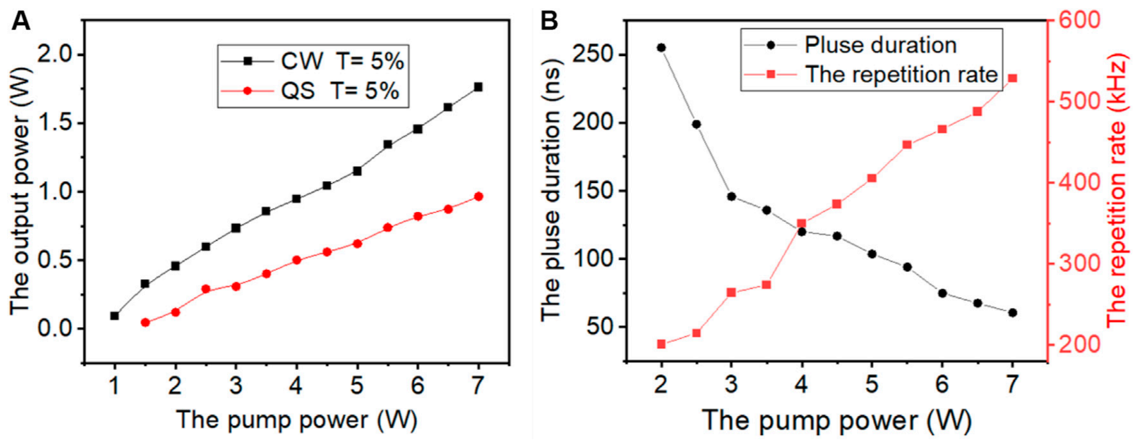

FIGURE 4
(A) Average output power. **(B)** Pulse duration and pulse repetition rate of the Ge-N SA-based Q-switched Nd:YVO_4 laser versus the pump power.

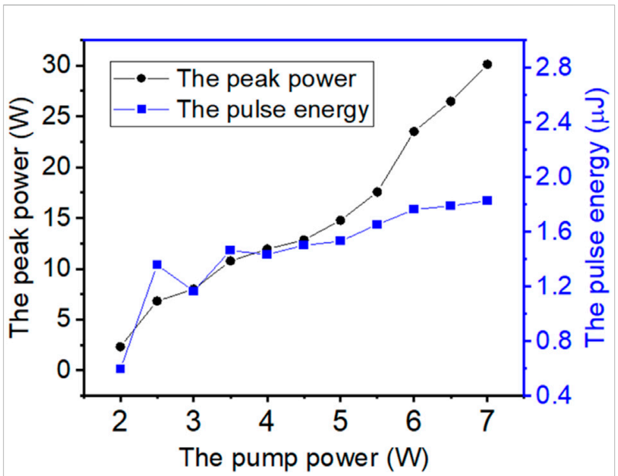

FIGURE 5
Peak powers and pulse energies of the Ge-N SA-based Q-switched Nd:YVO_4 laser versus the pump power.

Here, I and I_{sat} are the input intensity of the laser and saturation intensity of Ge-N SA, respectively, and ΔT is the modulation depth with a fitting value of 11.6%. The nonsaturable loss T_{ns} is about 12.2%.

Experimental setup and results

Experimental setup

Figure 3 shows the experimental setup for studying the lasing characteristics of Q-switched pulses using a passively Q-switched Nd:YVO_4 laser with Ge-N SA. To accomplish stable and effective Q-switching, a conventional 3-cm-long plane–plane cavity was used. A fiber-coupled laser diode (LD) emitting 808 nm was used as the pump source. Using an optical imaging system with a 1:1 imaging ratio, the pump beam with a spot radius of 200 μm was focused into a 3 mm^3 × 3 mm^3 × 8 mm^3 Nd:YVO_4 crystal. The Nd:YVO_4 crystal with the Nd-doping concentration of 0.5 at% was antireflection (AR) coated at 808 and 1,064 nm for a facet and AR coated at 1,064 nm for the other facet. As mature laser media for diode-pumped solid-state lasers, neodymium-doped crystals owe broad absorption bands and large emission cross sections as well as high environmental stability, which are beneficial to generate high peak power and narrow pulse width [39, 40]. M1 is a plane mirror with AR coated at 808 nm and high reflection (HR) coated at 1,064 nm. For the plane output coupler (OC) M2, the transmission (T) at 1,064 nm is 5%. The germanene SA was placed as close to the OC as possible to provide steady and effective Q-switching. A long-pass filter was also put behind the OC to reduce the residual pump power. The temporal pulse morphology was recorded using a digital oscilloscope (Tektronix DPO 4104B, United States) with a fast photo-detector (Thorlabs DET08 C/M, United States).

Experimental results and discussion

In the Nd:YVO_4 laser shown in Figure 3, the stable Q-switched laser oscillation based on Ge-N SA is realized when the pump power is increased above 1.8 W. As the pump power gradually increases, the output characteristics of the continuous wave (CW) and passively Q-switched pulses are measured.

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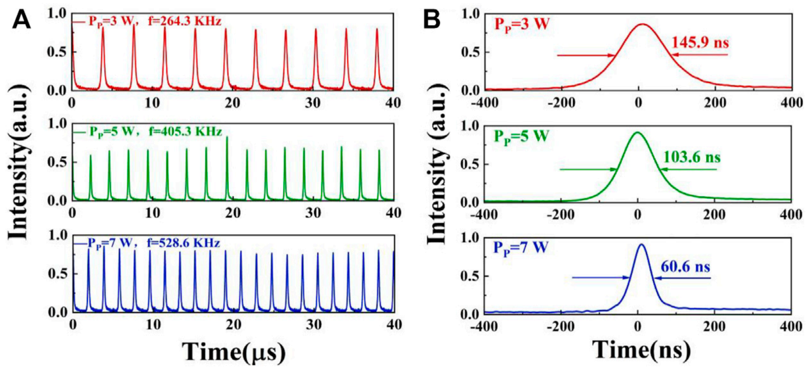

FIGURE 6
(A) Pulse sequence and **(B)** single pulse profiles of the passively Q-switched laser under the pump power of 3 W, 5 W, and 7 W.

TABLE 1 Passively Q-switching performances for the 1064 nm solid-state laser with different 2D SA materials.

SA type	Laser crystal	Pulse width (ns)	Repetition rate (kHz)	Pulse energy (μJ)	Peak power (W)	Ref
Graphene	Nd:YAG	400	850	0.68	1.69	[42]
BP	Nd:YAG	55	5,600	0.02	0.41	[43]
WS_2	$Nd:YVO_4$	98	392	0.441	4.5	[44]
WS_2	Nd:YAG	24	6,100	0.02	0.98	[43]
Antimonene	Nd:YAG	129	569.1	0.23	1.77	[45]
MoS_2/GaAs	$Nd:YVO_4$	51.3	769.7	0.417	8.14	[46]
Fe_3O_4	$Nd:YVO_4$	53	576.4	0.18	3.53	[47]
Carbon nanotube	Nd:YAG	1,200	95	4.5	3.75	[48]
Bi_2Te_3	Nd:YAG	576	28.57	5.24	9	[49]
Gold nanorods	Nd:YAG	3,100	50	19	6.13	[50]
Germanene	$Nd:YVO_4$	60.6	528.6	1.83	30.12	This work

The output power of CW and Q-switched pulses is shown in Figure 4A as a function of pump power. With the pump power of 7 W and the output coupler of 5%, the CW average output power is 1.762 W with the corresponding slope efficiency of 26%. Then, the Ge-N SA is put into the resonant cavity, and the average output powers of the passively Q-switched operation are recorded. At the pump power of 7 W, the maximum average output power of 0.965 W with the corresponding slope efficiency of 16.4% can be obtained. We have also been focusing on the state of Ge-N SA with increasing pump power. When the pump power reaches 7 W, the fluctuation of the average output power is measured at about 2.8%. By observation, there is no damage to Ge-N SA, and the distribution of Ge-Ns on the substrate seems to be uniform.

The pulse duration and repetition rate of the Ge-N-based Q-switched laser are measured and exhibited in Figure 4B. With the pump power ranging from 2 to 7 W, the pulse repetition rate gradually increases from 200.9 to 528.6 kHz, and the pulse width gradually narrows from 255 to 60.6 ns. This trend is consistent with the principle of passively Q-switched pulses [41].

The peak power and single pulse energy of passively Q-switched pulses are calculated by the measured pulse width, repetition rate, and average output power. Figure 5 shows the relationship between peak power and pulse energy and pump power. The maximum peak power of 30.12 W and single pulse energy of 1.83 μJ are obtained at the pump power of 7 W.

Figures 6A,B show the pulse train and single Q-switched pulse diagrams of the Ge-N SA-based Q-switched $Nd:YVO_4$ laser for a pump power of 2.5, 4.5, and 7 W, respectively. To further validate the stability of the Q-switching characteristics of germanene, the Q-switched laser based on Ge-Ns SA ran 6 h a day for 5 days under the pump power of 7 W. The Q-switched pulses keep high repeatability, and the amplitude fluctuation is less than 7%. By employing the 90.0/10.0 scanning-knife-edge method, the beam quality factor M^2 of the Q-switched laser based on Ge-N SA was also measured to further demonstrate the robustness of the Q-switching system. In the horizontal and longitudinal planes, the calculated M_x^2 and M_y^2 are 1.96 and 1.51, respectively.

Table 1 lists some key parameters of all-solid-state Q-switched laser using different 2D nanomaterials as SA at 1,064 nm. It can be found that the single pulse energy and pulse peak power produced by germanene SA are larger than those of most other two-dimensional materials. The results show that germanene has good nonlinear optical properties and is suitable for generating stable Q-switched pulses with high peak power in a solid-state laser.

Conclusion

In summary, a 4-nm-thick germanene nanosheet was fabricated successfully by the LPE method and applied as SA to generate a Q-switched pulse in $Nd:YVO_4$ all-solid-state lasers for the first time. The nonlinear optical parameters of the germanene nanosheet at 1,064 nm are measured by experimental methods. The stable Q-switching operation with large single pulse energy and high peak power is realized. The shortest pulse width of 60.6 ns was obtained with a maximum repetition rate of 528.6 kHz, and the associated single pulse energy and peak power are 1.83 μJ and 30.12 W, respectively, when the pump power is 7 W. The results indicate that germanene has excellent optical nonlinear properties and has broad application prospects in the field of all-solid-state laser.

Data availability statement

The original contributions presented in the study are included in the article/Supplementary Material; further inquiries can be directed to the corresponding authors.

Author contributions

BX and WT conceived and designed the experiments, performed the experiments and analyzed the data, and drafted the manuscript; WS and JW contributed to performing the theoretical analysis; XH and KJ fabricated and characterized the saturable absorber; WX provided experimental equipment, and all authors contributed to writing and editing the manuscript. All authors read and agreed to the published version of the manuscript.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Publisher's note

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