

Self-Q-switched and wavelength-tunable tungsten disulfide-based passively Q-switched Er:Y₂O₃ ceramic lasers

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We report on diode-pumped Er:Y₂O₃ ceramic lasers at about 2.7 μm in the tunable continuous-wave, self-Q-switching and tungsten disulfide (WS₂)-based passively Q-switching regimes. For stable self-Q-switched operation, the maximum output power reaches 106.6 mW under an absorbed power of 2.71 W. The shortest pulse width is measured to be about 1.39 μs at a repetition rate of 26.7 kHz at maximum output. Using a spin-coated WS₂ as a saturable absorber, a passively Q-switched Er:Y₂O₃ ceramic laser is also realized with a maximum average output power of 233.5 mW (for the first time, to the best of our knowledge). The shortest pulse width decreases to 0.72 μs at a corresponding repetition rate of 29.4 kHz, which leads to a pulse energy of 7.92 μJ and a peak power of 11.0 W. By inserting an undoped YAG thin plate as a Fabry–Perot etalon, for the passive Q switching, wavelength tunings are also demonstrated at around 2710, 2717, 2727, and 2740 nm. © 2018 Chinese Laser Press

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1. INTRODUCTION

In recent years, mid-infrared lasers near 3 μm have attracted great interest because of their important applications in remote-sensing technology, atmospheric-environment monitoring, and especially in biomedical science, due to their strong water absorption and the penetration depth of a few microns in biological tissue [1,2]. Er³⁺-doped materials make it possible to achieve 3-μm laser sources, which correspond to laser transition from ⁴I_{11/2} to ⁴I_{13/2} by pumping ground-state (⁴I_{15/2}) Er³⁺ ions to the ⁴I_{11/2} level using compact InGaAs laser diodes emitting around 970 nm [3–5]. The early research can be dated back to the 1960s, when Robinson and Devor first demonstrated a 2.69-μm stimulated emission in an erbium-doped mixed fluoride crystal [6]. After that, a variety of Er³⁺-doped laser materials have been developed based on fluoride and oxide hosts [2–10]. In recent years, sesquioxides, as desired host materials, have excited rising research interest for high-power laser sources because of their good thermomechanical property [11–13]. Moreover, rare-earth ions doped into sesquioxides could generate ultrafast lasers because of spectral broadening

of the optical transitions arising from large splitting of the ground state [14,15].

Pulsed lasers, rather than continuous-wave lasers, based on Q-switching and mode-locking technologies could have more potential in various applications. However, probably because there are no effective and easily available saturable absorbers, little early research concerning 2.8-μm Er³⁺ lasers has involved passively Q-switched and mode-locked operation. This situation is now changing with the development of 2D materials that have been reported to have advantages such as ultrabroadband saturable absorption, low cost, and easy fabrication. These merits have made 2D materials very popular for Q-switched and mode-locked laser operation from the visible to mid-infrared wavelength ranges [16–28]. In fact, in terms of passively Q-switched mid-infrared lasers as the investigated topic in this work, recently graphene [21], MoS₂ [22], tungsten disulfide (WS₂) [23], MoTe₂ [24], ReS₂ [25], Bi₂Te₃ [26], and black phosphorus (BP) [27,28] have been successfully used as saturable absorbers in the fields of solid-state lasers and/or fiber lasers. At present, WS₂-based mid-infrared laser operation has not yet been reported in a solid-state laser.

Transitional metal dichalcogenides (TMDs) are outstanding among 2D materials because of their superior properties of nonzero bandgap and layer-dependent third-order optical non-linearity. Recently, as a member of TMD family, WS_2 has been developed as a saturable absorber. Similar to MoS_2 , defects, boundary effects, and impurities of WS_2 could modify its bandgap structure, decreasing from 1.18 to 0.02 eV (61.6 μm) and 0.65 eV (1.89 μm) by suitably introducing W and S defects, respectively [23]. As a consequence, WS_2 should exhibit broadband saturable absorption properties, which deserves in-depth investigations.

Self- Q switching has been recognized as an interesting and effective measure for the generation of nanosecond-to-microsecond pulses with no special modulation elements to initiate and sustain the pulsing mechanism. At present, the investigation of self- Q switching has been mainly focused on near-infrared lasers. However, with respect to mid-infrared emission, it has seldom been studied. Recently, Liu *et al.* reported a self- Q -switched Er, Pr:CaF₂ laser at 2803.7 nm with a maximum output power of 262 mW; the pulse width was 718 ns [29]. A self-pulsed Er:Y₂O₃ ceramic laser has also been reported once with an average output power of 12 mW under single-end pumping geometry [30]. A more complicated laser configuration by using double-end pumping with two Er:Y₂O₃ ceramics increased the average output power to 83 mW [30]. Considering the significance of self-pulsed mid-infrared lasers and the state of the art, further investigations on this topic are still very necessary.

In this paper, stable self- Q switching has been investigated in a moderately doped Er:Y₂O₃ sesquioxide ceramic. Moreover, WS_2 has been developed for the first time as a saturable absorber for a passively Q -switched Er:Y₂O₃ laser. Additionally, by inserting an undoped YAG thin plate into the resonator, wavelength tunings of the Er:Y₂O₃ laser around 2710, 2717, 2727, and 2740 nm have also been demonstrated.

2. EXPERIMENTAL SETUP

Figure 1 shows the schematic of a diode-pumped Er:Y₂O₃ ceramic laser experimental setup. The pump source is a fiber coupled InGaAs diode laser with a core diameter of 105 μm and a numerical aperture of 0.22. The peak emitting wavelength of the diode laser is about 976 nm at a maximum output power of about 25 W at room temperature. The pump beam was focused into the Er:Y₂O₃ ceramic by two doublet lenses having focal lengths of 30 and 50 mm. Thus, the waist size of the pump beam was expanded to about 175 μm , which

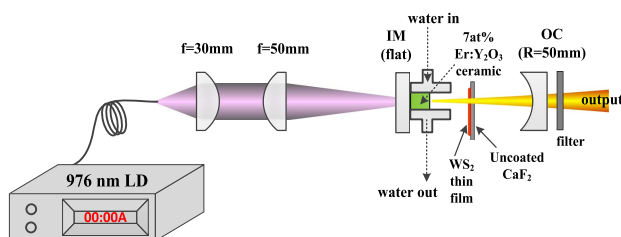


Fig. 1. Schematic of diode-pumped Er:Y₂O₃ ceramic lasers. IM, input mirror; OC, output coupler.

aimed at thermal mitigation of the laser gain medium. The flat input mirror has a transmission of about 95% at the pump wavelength and a high reflection of more than 99.9% at 2.7 μm . The 50-mm (curvature radius) output coupler has partial transmission of about 4.8% at 2.7 μm . The physical length of the laser resonator has been maintained to be about 50 mm during the experiments.

The laser gain medium was an Er:Y₂O₃ ceramic with a dopant concentration of 7 at. % and dimensions of $\phi 6 \times 7 \text{ mm}^3$. The Er:Y₂O₃ ceramic was wrapped with a piece of indium foil and then mounted inside a water-cooled copper block with the temperature set at 13°C. The Er:Y₂O₃ absorbed about 87% of the incident pump power. An undoped YAG thin plate was used to modulate the intracavity loss for wavelength tuning. For passive Q -switching, few-layer WS_2 thin film was fabricated and transferred onto a CaF₂ substrate serving as a saturable absorber.

3. RESULTS AND DISCUSSION

In free-running mode, namely, without inserting any intracavity elements, at laser threshold the lasing behavior was observed to show a self-pulsing phenomenon only by misaligning the laser resonator. Increasing the absorbed power to about 1.46 W, stable self- Q switching was found. Figure 2 shows the dependence of average output power on absorbed power of the stable self- Q -switched Er:Y₂O₃ ceramic laser. The maximum output power reached 106.6 mW at an absorbed power of about 2.72 W. Linearly fitting the data in Fig. 2 led to a slope efficiency of about 8.2%. At the maximum output, the pulse-to-pulse amplitude fluctuation of the self- Q -switched pulse train was measured to be about $\pm 9\%$. The laser spectrum measured at maximum average output power is shown as an inset in Fig. 2, which exhibits a two-peak structure with a main peak at 2717.02 nm and a side peak at 2709.9 nm. The main peak has an full width at half-maximum (FWHM) of about 2.18 nm. It should be pointed out that the dual-wavelength lasing behavior can only be observed when the average output power is close to the maximum. At threshold and low output power, only the single wavelength of 2717 nm can be found. It should be

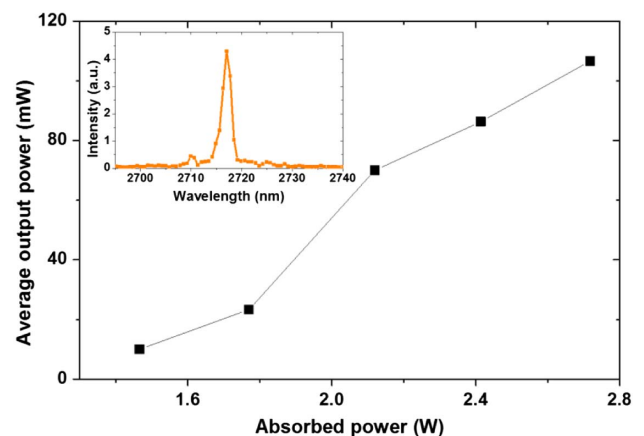


Fig. 2. Dependence of average output power on absorbed power of self- Q -switched Er:Y₂O₃ ceramic lasers; inset, corresponding laser spectrum at the highest output power.

pointed out that this result is far better than that reported in Ref. [30], with a maximum average output power of 10 mW in single-end pumping of an Er:Y₂O₃ ceramic laser. The self-pulsing phenomenon could be explained by the reabsorption effect of the Er:Y₂O₃ ceramic. As we know, lasing and spontaneous emission lead to considerable population of the ⁴I_{13/2} lower level. The ⁴I_{13/2} population reabsorbs the photons at the transition identical to that of laser radiation only if suitable upper and lower level lifetimes of the Er:Y₂O₃ ceramic can be satisfied [29].

Further increasing the absorbed power led to stability degradation of the self-*Q*-switching pulse trains. The pulse trains became disordered and finally degenerated to continuous-wave operation. This conversion from self-*Q* switching to continuous wave has also been observed in some other self-*Q*-switched lasers, e.g., Yb:CGB lasers [30,31]. To interpret this conversion phenomenon, the concentration-dependent upconversion process should be taken into account, which circumvents the well-known self-terminating effect and therefore makes it possible to lase 3- μ m Er³⁺ laser radiation. However, it is also clear that increasing absorbed power will finally result in the domination of the upconversion process in contrast to the reabsorption process, which causes continuous-wave operation instead of self-pulsing, eventually. From this point of view, it seems reasonable that we have obtained higher average output power than that reported in Ref. [30]. We have used a relatively weakly doped Er:Y₂O₃ material (7% versus 15%), which is of advantage to reducing upconversion.

Figure 3 shows the typical pulse trains and the corresponding single-pulse profiles at different output powers. At the threshold, the pulse width was measured to be about 1.61 μ s at a pulse repetition rate of 16.6 kHz, as shown in Fig. 3(a). At maximum output of the stable self-*Q*-switched laser operation, the pulse width reduced to about 1.39 μ s, and the corresponding pulse repetition rate increased to 26.7 kHz, as shown in Fig. 3(b). Figure 3(c) shows a typical shape and stability-degraded pulse trains with a repetition rate of 29.1 kHz, while the pulse width was found to expand to about 1.73 μ s. In fact, when the absorbed power was increased to about 3.7 W, the pulses completely collapsed, and at the same time, the output power

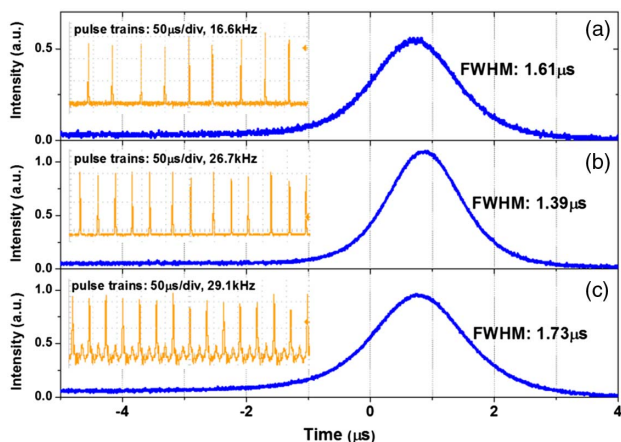


Fig. 3. Typical self-*Q*-switched single-pulse profiles with corresponding pulse trains as insets.

diminished greatly. Afterward, by aligning and optimizing the laser resonator, continuous-wave laser operation can be achieved with remarkable power improvement.

A passively *Q*-switched Er:Y₂O₃ ceramic laser was performed with a WS₂ saturable absorber. The few-layer WS₂ nanosheets were prepared by the liquid phase exfoliation method. The bulk WS₂ (200 mg) was sonicated for 20 h in N-2-methyl pyrrolidone (NMP, 200 mL) to produce the few-layer WS₂ suspension. The characterization of the few-layer WS₂ nanosheets is shown in Fig. 4. The bulk WS₂ and the few-layer WS₂ nanosheets are both characterized by X-ray diffraction (XRD), as shown in Fig. 4(a), in which all the labeled peaks of the bulk WS₂ correspond to hexagonal WS₂ (JCPDs NO. 08-0237). The XRD pattern of the few-layer WS₂ nanosheets only retained the *c* axis of the peaks, such as (002), (004), (006), and (008), and some characteristic peaks disappeared, which indicated the bulk WS₂ had been successfully exfoliated. Further, the Raman spectrum in Fig. 4(b) also confirmed the exfoliation of WS₂. The few-layer WS₂ nanosheets show E_{2g}¹ and A_{1g}¹ phonon modes at 348.8 and 416.6 cm⁻¹, respectively. These values are different from bulk WS₂ (351.9 and 417.1 cm⁻¹, respectively). The mode of few-layer WS₂ redshifts compared to that of bulk WS₂, which could be attributed to the phonon softening. Besides, in Fig. 4(c), an atomic force microscopy (AFM) image was given to characterize the thickness of the as-prepared few-layer WS₂. In Fig. 4(d), the height profile indicates the as-prepared WS₂ nanosheets are around 5.52 nm, which suggests the nanosheets are about five layers, since a WS₂ monolayer is ~1 nm [32].

The few-layer WS₂ suspension was then transferred onto a CaF₂ substrate by a simple and commonly used spin-coating method that involves dripping and rotating the WS₂ suspension onto the substrate. After drying the WS₂ suspension inside an oven with a constant temperature of 60°C for 5 h, WS₂ thin film can be formed for usage as a saturable absorber. In Fig. 5, the transmission of the blank CaF₂ substrate was measured to be about 94.5% at 2.7 μ m, which corresponds to the Fresnel

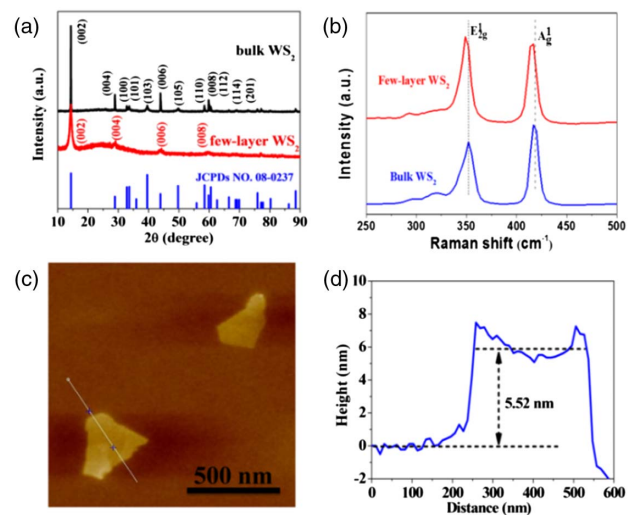


Fig. 4. (a) XRD patterns and (b) Raman spectra of the bulk WS₂ and few-layer WS₂ samples; (c) AFM image and (d) height profile of an as-prepared few-layer WS₂ sample.

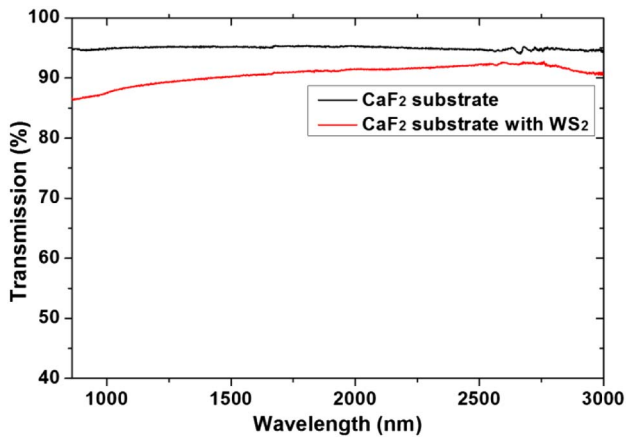


Fig. 5. Transmissions of the blank CaF₂ substrate and CaF₂ substrate with WS₂ thin film.

reflection loss. The transmission indicates the transparency and good quality of the CaF₂ substrate. The as-prepared WS₂ saturable absorber, namely WS₂ thin film plus the CaF₂ substrate, was also measured to have a transmission of about 92.2% at 2.7 μm. Thus, the final transmission of the WS₂ thin film could be deduced to be around 97.5%, which means a linear loss of about 2.5%. Moreover, a broad and flat transmission in the whole near- to mid-infrared spectral range can also be found from Fig. 5, which reveals the superiority of broadband saturable absorption of the WS₂ as a saturable absorber.

The saturable absorption intensity of the WS₂ was also measured at 1960 nm with a substitute Tm³⁺-doped mode-locked fiber laser with a pulse width of 680 fs, since a suitable 2.7-μm pulsed laser source is unavailable in our lab. In fact, according to the bandgap structure of WS₂, it should have lower saturable intensity at 2.7 μm than at 2.0 μm. Figure 6 shows the measuring result with fitted saturable intensity of 10.6 MW/cm², modulation depth of 6.7%, and nonsaturation loss of 6.4%.

Figure 7 shows the dependence of the average output power on the absorbed power of a WS₂-based Q-switched Er:Y₂O₃ laser with good stability. The laser threshold increased to about 3.1 W of absorbed power, which indicated a higher intracavity loss originating from the introduction of the WS₂ saturable

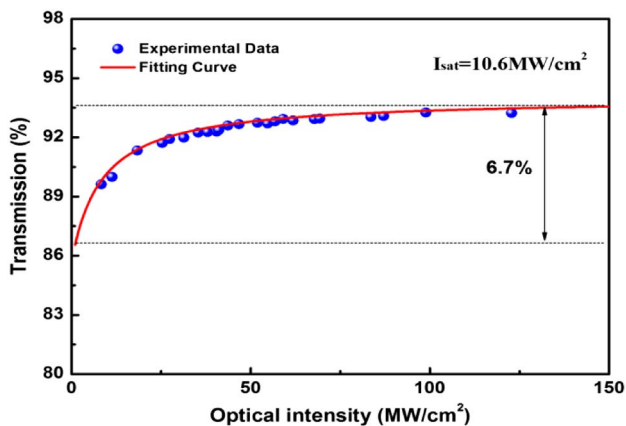


Fig. 6. Saturable absorption of the WS₂ saturable absorber used.

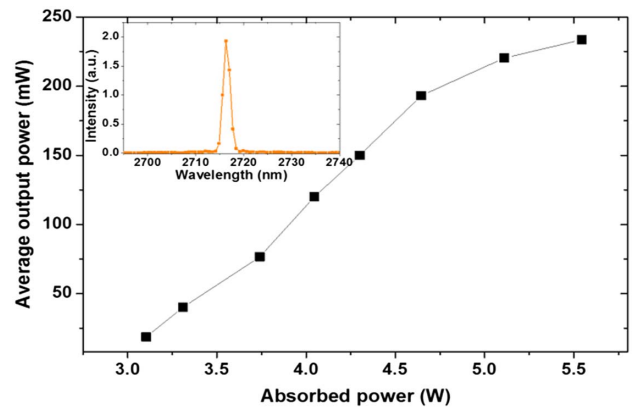


Fig. 7. Dependence of average output power on absorbed power of WS₂-based Q-switched Er:Y₂O₃ ceramic lasers; inset, corresponding laser spectrum at the highest output power.

absorber. The maximum average output power was found to be about 233.5 mW at an absorbed power of 5.54 W. Linearly fitting the data in Fig. 6 led to a slope efficiency of about 10.6%. At this maximum output, we measured the pulse-to-pulse amplitude fluctuation of the passively Q-switched pulse train to be about ±6%. We noticed that at this absorbed power, continuous-wave output power reached close to 600 mW. Optimizing the quality of the WS₂ thin film and depositing the CaF₂ substrate with antireflection coating for reducing the Fresnel loss could be resorted to for power scaling, which will be studied in our near future work. Further increasing the absorbed power led to the Q-switched pulse trains showing an instability trend. The stability degradation should arise from heat accumulation of the WS₂ thin film. The laser spectrum, showing a single-peak structure, peaks at 2716.3 nm with an FWHM of about 1.78 nm (see Fig. 7). That is to say, compared with the case of self-Q switching, the laser spectrum of passive Q switching is narrowed by 0.4 nm, which should be attributed to the etalon effect of the CaF₂ substrate.

Figure 8 shows the single-pulse profile with an FWHM value of about 0.72 μs at maximum output power. The pulse width narrowed to half the value of that of self-Q switching, which probably indicated that the modulation arising from

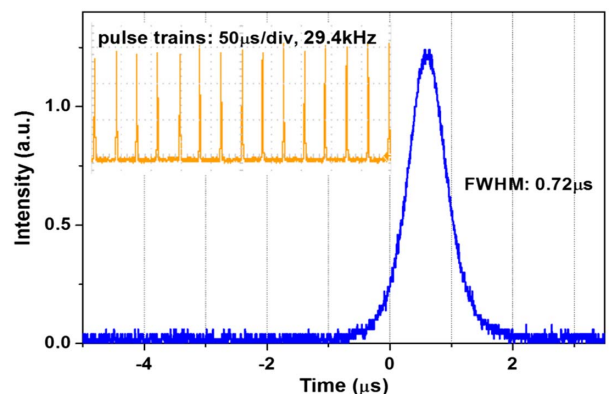


Fig. 8. Typical single-pulse profile of a passively Q-switched Er:Y₂O₃ ceramic laser; inset, corresponding pulse trains.

WS₂ is larger than that arising from reabsorption. Moreover, it is also possible that the reabsorption modulation was also introduced during the passive *Q* switching. In addition, one can see that the passively *Q*-switched laser operated at higher output than did self-*Q* switching with higher absorbed power, which is also helpful in producing pulses with narrower width. At maximum output, stabilities of average output power and pulse temporal profile were both measured in 1 h, as shown in Fig. 9. The stability of average output power was found to be about 4.7% with respect to the maximum average output power. For the temporal pulse profile, it has been estimated to have a time jitter of about ± 0.09 μ s. The recorded 0.72- μ s pulse corresponded to the shortest pulse time duration.

At a large scale, the pulse trains can also be observed to have a repetition rate of 29.4 kHz. Figures 10(a) and 10(b) show the

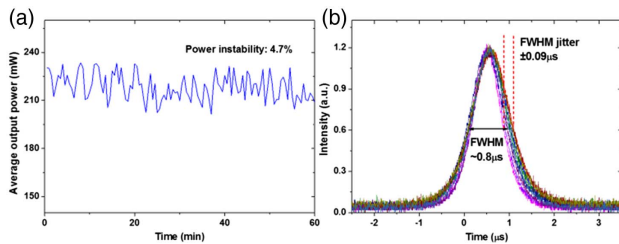


Fig. 9. (a) Stability measurement of average output power in 1 h and (b) temporal profiles of 12 superimposed pulses recorded every 5 min in 1 h.

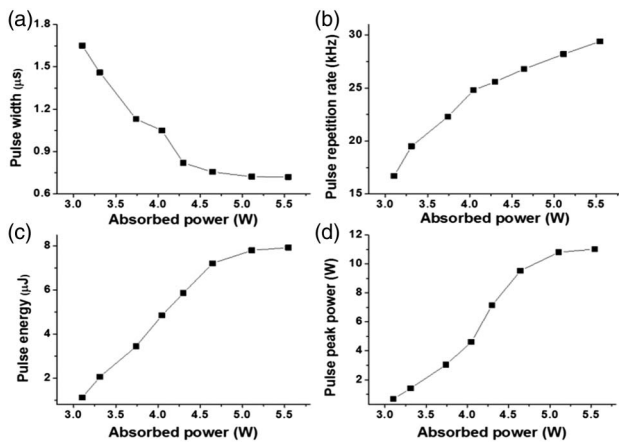


Fig. 10. Dependences of (a) pulse width, (b) pulse repetition rate, (c) pulse energy, and (d) pulse peak power on absorbed powers.

entire evolutions of pulse width and repetition rate with the increase of the absorbed powers. Basically, the pulse width shows monotonous decrease as the absorbed power increases. However, it is also very obvious that the trend of pulse narrowing is weakening. In particular, the pulse width shows saturation when reaching an absorbed power of 4.5 W. As for the pulse repetition rate, it increased from 16.7 to 29.4 kHz, showing about three-segment linear increases with decreasing slopes with the increase of the absorbed power. Knowing the pulse width and repetition rate, we further estimated the values of pulse energy and pulse peak power of the passively *Q*-switched Er:Y₂O₃ laser, as shown in Figs. 10(c) and 10(d). The pulse energy increased from 1.11 to 7.92 μ J, while at the same time the pulse peak power increased from 0.67 to 11.0 W.

In Table 1, we compared the results of some recent research in regard to diode-pumped passively *Q*-switched mid-infrared solid-state lasers based on 2D material saturable absorbers. From Table 1, one can see that the present result is indeed pretty good, with relatively large output power and pulse energy. Therefore, it could be safely concluded that WS₂ as a saturable absorber applicable for mid-infrared wavelength is quite competitive with other 2D materials, and it is worth developing further.

According to the emission spectrum of Er:Y₂O₃ ceramic [33], 2717-nm emission dominates with the highest emission intensity. However, one should also notice that there are several narrow emission bands besides the 2717-nm peak, which leaves room for realizing laser operation at other emission bands. Finally, wavelength tuning was realized by adjusting the laser resonator with the help of a 0.13-mm-thick pure YAG crystal acting as a Fabry–Perot etalon with a free spectral range (FSR) of about 634 GHz. Compared with CaF₂ material, undoped YAG has a relatively larger refractive index, which is believed to produce larger transmission differences (up to about 29% instead of 7% for CaF₂) among the potential lasing wavelengths. Thus, it could allow the possibility of favoring low gain lines over high gain lines. Consequently, emission lines with lower gains could lase, which will lead to broader wavelength tuning.

The achieved laser emissions in the passively *Q*-switched regime are registered in Fig. 11. By slightly tilting the YAG thin plate, laser emissions around 2710, 2717, 2727, and 2740 nm have been successfully operated with a total wavelength range of about 8 nm. Some dual-wavelength laser emissions can be obtained, as shown in Fig. 11. The disconnected wavelength tuning was mainly limited by the narrow emission bands of the Er:Y₂O₃ ceramic. On the other hand, the present tuned wavelengths could indicate that mode locking is probably realizable in

Table 1. Diode-Pumped Passively *Q*-Switched Mid-Infrared Solid-State Lasers Based on 2D Material Saturable Absorbers

2D Material	Laser Material	Max Output Power	Min Pulse Width	Max Pulse Energy	Ref.
MoS ₂	Er:Lu ₂ O ₃	1.03 W	335 ns	8.5 μ J	[22]
MoTe ₂	Ho, Pr:LiLuF ₄	73 mW	670 ns	0.95 μ J	[24]
ReS ₂	Er:SrF ₂	0.58 W	508 ns	12.1 μ J	[25]
BP	Er:SrF ₂	180 mW	702 ns	2.34 μ J	[28]
Graphene	Ho, Pr:LLF	88 mW	937.5 ns	1.6 μ J	[34]
WS ₂	Er:Y ₂ O ₃	233.5 mW	0.72 μ s	7.92 μ J	This work

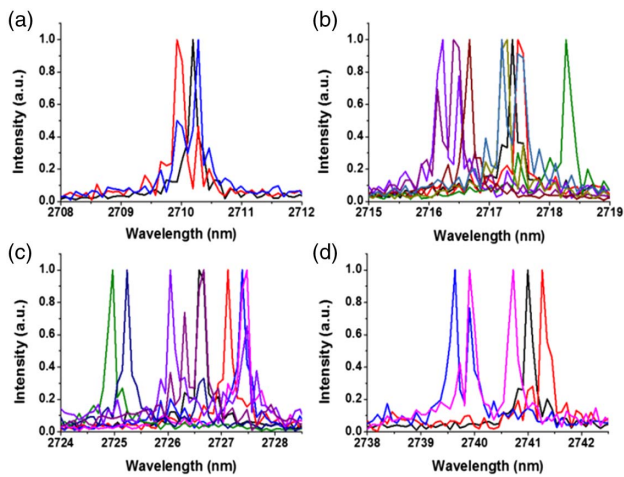


Fig. 11. Wavelength tunings of the Er:Y₂O₃ ceramic laser emissions around 2710, 2717, 2727, and 2740 nm.

Er:Y₂O₃ ceramic by configuring a laser resonator with large length.

4. CONCLUSION

In conclusion, tunable continuous-wave, self-*Q*-switched and WS₂-based passively *Q*-switched Er:Y₂O₃ ceramic lasers at about 2.7 μm have been realized. As for self-*Q*-switched operation, a maximum output power up to 106.6 mW was achieved with the shortest pulse width of about 1.39 μs at a repetition rate of 26.7 kHz. A passively *Q*-switched Er:Y₂O₃ laser has been further realized with a maximum average output power of 233.5 mW. The shortest pulse width is about 0.72 μs at a corresponding repetition rate of 29.4 kHz, which leads to a pulse energy of 7.92 μJ and a peak power of 11.0 W. Finally, wavelength tunings have also been demonstrated at around 2710, 2717, 2727, and 2740 nm by inserting an un-doped YAG etalon into the laser resonator. In the near future, further power scaling of the passively *Q*-switched Er:Y₂O₃ ceramic laser could be expected by optimizing the quality of the WS₂ saturable absorber and by coating the CaF₂ substrate. Narrowing the pulse width for improving the pulse energy and peak power could also be achieved at the same time.

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