A Single-Longitudinal-Mode Passively Q-Switched Nd:YVO_4 Laser Using Black Phosphorus Saturable Absorber *

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(Received 17 October 2016)

A black phosphorus (BP) saturable absorber (SA) solution with different concentrations (1.0 and 0.5 mg/ml) is fabricated with the liquid-phase exfoliation method. By using the BP-SA, a compact diode-pumped passively Q-switched Nd:YVO_4 laser is demonstrated. One reflecting Bragg gratings is used as the output coupler for mode selection. By inserting those BP-SA solutions in the laser cavity, the maximum single longitude mode, Q-switched output powers of 126 mW at 692.5 kHz and 149 mW at 630.3 kHz are achieved at the pump power of 8.0 W, corresponding to the pulse durations of 144 ns and 196 ns, respectively. Moreover, longitudinal-mode characteristics of Q-switched output laser in different optical cavity lengths based on two-kind BP-SA solution concentrations are investigated. Our results show that BP-SA could also be developed as an effective SA for the Q-switched, single longitudinal mode pulse laser.

PACS: 42.55.Xi, 42.60.Gd, 42.70.Hj DOI: 10.1088/0256-307X/34/1/014204

Two-dimensional (2D) nanomaterials have attracted much attention recently due to their remarkable electronic and optical properties, especially the emergence of graphene and graphene-like 2D nanomaterials due to their unique zero bandgap, high thermal stability, the excellent saturable absorption properties, and easy fabrication.[1–3] Nevertheless, it provides unprecedented chances for producing novel and distinctive electronic and optoelectronic devices. With the wide application of 2D nano-materials SA, other graphene and graphene-like 2D materials have also attracted extensive attention.

Recently, as a new type of 2D nano-material, BP has attracted great interests in various areas of electronics and photonics,[4–6] owing to its tunable direct bandgap,[7] high carrier mobility ($\sim$1000 $cm^2/V{\cdot}s$),[8] large on/off ratios (>105),[9] and high in-plane anisotropy[10,11] at room temperature. These characteristics give BP high absorption, adjustable optical modulation depth and ultrafast recovery time property. Moreover, BP's direct bandgap can be tuned from 0.35 eV (bulk) to 2.0 eV (monolayer),[12,13] which fills the present gap between the zero bandgap of graphene and wide bandgap of semiconducting transition metal dichalcogenides. Thus it can be capable of a broadband optical response with a wavelength range from $\sim$0.6 μm to $\sim$4 μm, depending on the number of layers of phosphorus. These features may lead to wide-range potential photonics devices, such as the Q-switcher for pulse lasers in a wide spectral range from visible to infrared.

As a promising application, BP can be used as SAs for the generation of short pulses in Q-switched or mode-locked lasers, benefitting from the features of high absorption, adjustable optical modulation depth and ultrafast recovery time. BP Q-switched solid state lasers have been realized at the pulse durations of 620 ns, 1.73 μs, 189 ns, 189 ns, and the corresponding maximum pulse energies are 326 nJ, 90 nJ, 104 nJ and 205 nJ, respectively.[14–17] Furthermore, 1.18 μs pulses with 7.7 μJ, 2.96 μs pulses with 194 nJ, 2.53 μs pulses with 276 nJ, and 10.32 μs pulses with 94.3 nJ were obtained from the BP-SAs passively Q-switched Er-doped fiber.[18–21] In comparison with fiber lasers, solid-state lasers possess superiorities such as further pulse energy scaling, pulse duration narrowing and mode selection.

Extending the BP-SAs passively Q-switched laser to the single longitudinal mode operation remains an interesting topic. Reflecting Bragg grating (RBG) in photo-thermo-refractive (PTR) glass has versatile and unique optical properties, such as perfect thermal stability, narrow diffraction spectrum, good angular selectivity, flexible diffraction efficiency, large transparent range, low losses and high laser damage threshold,[22–24] which have successfully paved the way for spectral narrowing and mode selection. Thus using one RBG as the BP-SA passively Q-switched laser output coupler can obviously reduce the laser spectral width.

In this Letter, we demonstrated that BP-SA fabricated by the liquid phase exfoliation method can be used as an SA to generate short pulses in Nd:YVO_4 lasers with an RBG as the output coupler. Compared with solid state absorbers, liquid absorbers have higher heat dissipation, and no damage happens at the contact between the two interfaces. Two kinds of BP-SA solution with concentrations of 1.0 and 0.5 mg/ml were injected into quartz cells. After that, the quartz cells were inserted into a Nd:YVO_4 laser cavity, re-

*Supported by West Young Scholar Foundation of the Chinese Academy of Sciences under Grant No XAB2015B27.

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spectively. Two single longitudinal mode pulses with the minimum pulse durations of 144 ns at 692.5 kHz and 196 ns at 630.3 kHz, the maximum average output powers of 126 mW and 149 mW were achieved correspondingly. It is verified that the shorter pulse duration and the higher repetition rate can be achieved by the higher concentration of BP-SA solution.

In the experiment the few-layer BP nanoflakes were fabricated by the liquid-phase exfoliation technique.[24] Firstly, the BP powder was exfoliated and dispersed into N-methyl-2-pyrrolidone (NMP) solvent and ultra-sonicated for 6 h. To remove the large-size phosphorous sheets, as-prepared few-layer BP was centrifuged at 6000 rpm for 6 min to remove the large number of thick flakes. After that, the obtained supernatant liquor was collected for further use. The optical absorption property of the prepared BP-SA was characterized using an optical spectrometer. The measured transmission spectrum of BP-SA solution with concentrations of 1.0 and 0.5 mg/ml from 800 to 1100 nm is illustrated in Fig. 1(a), which shows that the optical transmittances of the quartz cell filled with BP-NMP solution are 87.7% and 92.3% at 1064 nm, respectively. The Raman spectrum of the as-prepared BP-SA solution with the concentration of 1.0 mg/ml was measured with a Raman spectrometer excited by a 532 nm laser source, as illustrated in Fig. 1(b). Three distinct Raman peaks at 356.4 cm^{-1}, 429.4 cm^{-1} and 456.8 cm^{-1} are clearly exhibited, corresponding to the A_g^1 (out-of-plane), B_{2g} and A_g^2 (in-plane) vibration modes of the layered BP, respectively.

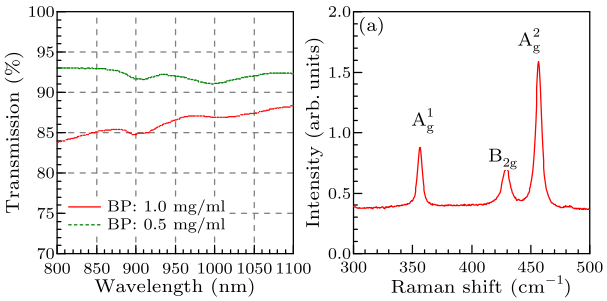

Fig. 1. (a) Transmission spectra of BP-SA with different concentrations. (b) Raman spectra of BP-SA at room temperature.

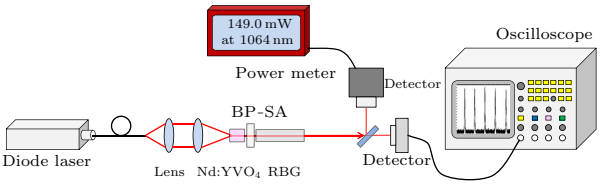

Fig. 2. Schematic configuration of the Q-switched $Nd:YVO_4$ laser.

The experimental arrangement for the BP-SA Q-switching of a diode-pumped $Nd:YVO_4$ laser is schematically shown in Fig. 2. The pump source was a fiber-coupled 808 nm laser diode with a core diameter of 200 μm and numerical aperture of 0.22. Its radiation was coupled into the laser crystal, by using a couple of convex lenses (1:1). The a-cut $Nd:YVO_4$ (0.5 at.% of Nd concentration) used in the experiment has a length of 5 mm and a square aperture of $3.0 \times 3.0\,mm^2$. The $Nd:YVO_4$ was coated for high reflection at the lasing wavelength of 1064 nm and for anti-reflection at the pump wavelength of 808 nm on the side towards the pump. The other surface of $Nd:YVO_4$ was antireflection coating at the pump and laser wavelengths. The $Nd:YVO_4$ was wrapped with an indium foil and then mounted on a water-cooled copper crystal holder with the cooling water temperature of 20°C to preserve the laser crystal from thermal fracture. The BP-SA solution with concentrations of 1.0 and 0.5 mg/ml in a 1-mm-thick quartz cell is used as the SA, respectively. Spectral narrowing and mode selection were achieved by replacing the laser output coupler by an RBG (Optigrate Corp.). The RBG was fabricated by holographic recording inside photothermo-refractive (PTR) glass. The spectral dependence of diffraction efficiency of the RBG used in our experiments was centered at the wavelength of 1064.04 nm. The maximum diffraction efficiency of RBG was 31%. The thickness of RBG was 20 mm with a clear aperture of $3.0 \times 3.0\,mm^2$. The output power was detected using a power meter (PM206 Power Meter, Throlabs Inc.), and the repetition rate and pulse duration were recorded using a digital oscilloscope (Tektronix DPO 4104, Tektronix Inc.) with a free-space InGaAs detector (DET08C, Throlabs Inc.), as shown in Fig. 1. Meanwhile, the laser spectrum and longitudinal mode characteristics of laser output were detected using a spectroscope (YOKOGAWA AQ6370D) and a Fabry–Perot interferometer, respectively. The free spectral range of the Fabry–Perot interferometer was 10 pm and the resolution ratio was 0.2 pm.

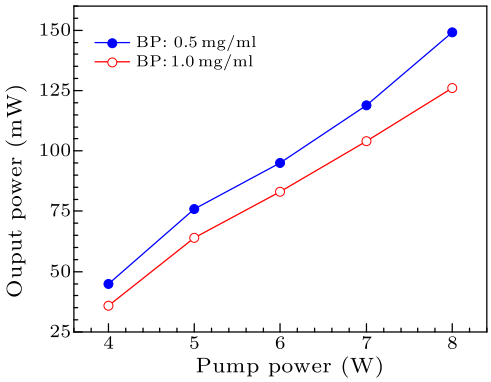

Fig. 3. Average output power and pulse energy versus the pump power.

Firstly, the laser performance using the RBG as the output coupler with different concentrations of BP-SA solution was investigated. The laser was found to operate in the Q-switch mode. However, the absorbed pumped power rose up to 4.0 W, the Q switching was initiated with BP-SA. In Fig. 3, with the BP-

SA solution with concentrations of 1.0 and 0.5 mg/ml, the maximum average output powers were 126 mW and 149 mW with an absorbed pump power of 8.0 W, corresponding to the optical-to-optical conversions of 1.575% and 1.8625%, respectively. This low output power was attributed to a large linear loss of the BP-SA. In comparison of the two curves in Fig. 3, we could obviously see that the higher the concentration of BP-SA solution, the lower the average output power. Further measurement indicated that the passively Q-switched Nd:YVO_4 laser output was polarized along the c principal axis of the Nd:YVO_4 crystal. The polarization extinction ratio was about 100:1.

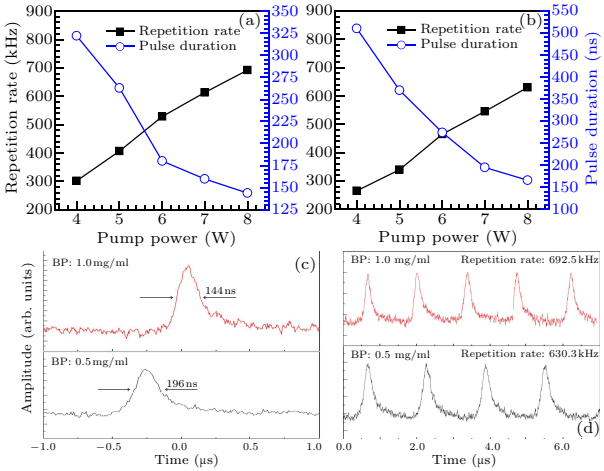

Fig. 4. (a) The repetition rate and the pulse duration versus the pump power of the Q-switched laser based on BP-SA solution with the concentration of 1.0 mg/ml. (b) The repetition rate and the pulse duration versus the pump power of the Q-switched laser based on BP-SA solution with the concentration of 0.5 mg/ml. (c) Single pulse profile of the Q-switched laser with the BP-SA solution concentrations of 1.0 and 0.5 mg/ml. (d) A typical Q-switched pulse train with the BP-SA solution concentrations of 1.0 and 0.5 mg/ml.

Figures 4(a) and 4(b) plot the evolutions of the repetition rate and pulse duration with the pump power. Similar to other passively Q-switched lasers, the higher the pump intensity was, the easier the BP-SA was bleached and the shorter the pulse duration was. Thus we expect to see the narrower pulse duration and larger pulse repetition rate with the increasing pump power. Consequently, for the pump power of 8.0 W, the pulse durations were 144 ns at 692.5 kHz repetition rate and 196 ns at 630.3 kHz repetition rate with the BP-SA solution concentrations of 1.0 and 0.5 mg/ml, respectively. The measured repetition rate and pulse duration are shown in Fig. 4(a). Figures 5(a) and 5(b) show the temporal pulse profile and the typical Q-switched pulse train, in which the pulse-to-pulse instability is below 10%. Additionally, because the thermal loading on the surface of BP-SA could be dissipated rapidly in the NMP solvent, the BP-SA still kept its effectiveness and any damage on the SA has not be observed. Therefore, the results demonstrated that the BP-SA solution possesses adjustable optical absorption, high damage threshold, and high heat dissipation and can be used for a high power passive Q-switch operation.

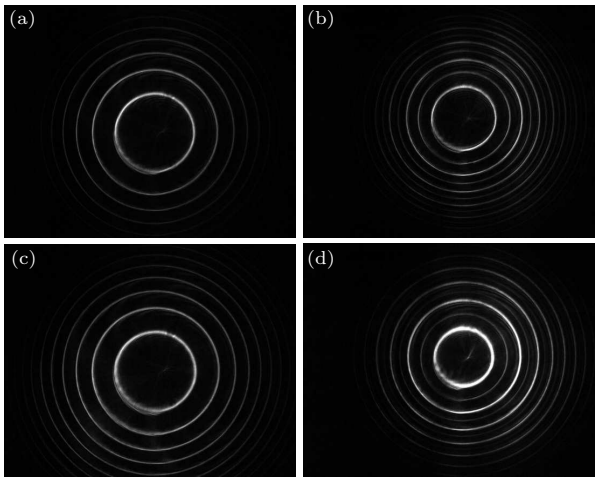

Fig. 5. (a) The spectra of the laser output with the concentration of 1.0 mg/ml BP-SA at single longitudinal mode operation when the optical cavity lengths are 27 mm and 40 mm, respectively. (b) The spectra of the laser output with the concentration of 1.0 mg/ml BP-SA at multi-longitudinal mode operation when the optical cavity length is 54 mm. (c) The spectra of the laser output with the concentration of 0.5 mg/ml BP-SA at single longitudinal mode operation when the optical cavity lengths are 27 mm, 40 mm and 54 mm, respectively. (d) The spectra of the laser output the concentration of 0.5 mg/ml BP-SA at multi-longitudinal mode operation when the optical cavity length is 81 mm.

Table 1. The longitudinal mode characteristics of the laser in different optical cavity lengths.

Optical cavity length (mm)	Longitudinal mode spacing (nm)	Results of laser output	
		BP-SA: 1.0 mg/ml	BP-SA: 0.5 mg/ml
27	0.021	Single longitudinal mode	Single longitudinal mode
40	0.014	Single longitudinal mode	Single longitudinal mode
54	0.0105	Multi-longitudinal modes	Single longitudinal mode
81	0.007	Multi-longitudinal modes	Multi-longitudinal modes

Aiming at further studying the longitudinal mode characteristics of the Q-switched laser output, we consider the laser operation with BP-SA solution concentrations of 1.0 and 0.5 mg/ml in different optical cavity lengths by the Fabry–Perot interferometer. We adjusted the position of RBG to obtain different optical cavity lengths and the results measured are listed in Table 1. As we can see from Table 1, a short optical cavity was characterized by the wide longitudinal mode spacing. It was required for longitudinal mode

selection to need the optical cavity length as short as possible. It is noteworthy that the physical thickness of RBG is thicker than the ordinary broadband dielectric mirror. The relationship between the physical thickness of RBG and the position of the virtual mirror could be calculated as in Ref. [26].

For the BP-SA solution concentration of 1.0 mg/ml, the laser operated at single longitudinal mode when the optical cavity lengths are 27 mm and 40 mm in Fig. 5(a). After that, the optical length increases to 54 mm, the rings split into two sub-rings as shown in Fig. 5(b). This split was produced by the multi-longitudinal modes of the laser. For the BP-SA solution concentration of 0.5 mg/ml, it is clear that the laser operated at single longitudinal mode when the optical cavity lengths are 27 mm, 40 mm and 54 mm as shown in Fig. 5(c). When the optical cavity length is up to 81 mm, the sub-rings observed directly in Fig. 5(d) arise from the individual longitudinal modes of the laser. Under this condition, the laser output is composed of multi-longitudinal modes.

Compared with the Q-switch operation with the BP-SA solution concentration of 1.0 mg/ml, the cavity length of the Q-switched laser that could provide single longitudinal mode is obviously shorter. It is demonstrated that the laser longitudinal mode selection ability would be reduced by higher concentration of BP-SA in the same optical cavity length. Since a higher concentration of BP-SA made less round trips between two reflecting surfaces for the development of a pulse, it is important for the single longitudinal mode selection to need round trips as many as possible.

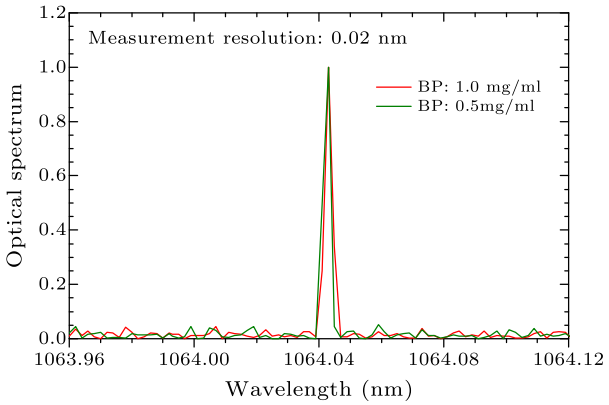

Fig. 6. Optical spectrum of the single longitudinal mode laser.

The optical spectrum of the Q-switched laser based on the BP-SA solution concentrations of 1.0 and 0.5 mg/ml was measured at the maximum output power, as shown in Fig. 6. The resolution of the optical spectrum analyzer is 0.02 nm and the peak wavelength is 1064.043 nm.

In summary, a Q-switched Nd:YVO_4 laser based on the two-kind BP-SA solution with concentrations of 1.0 and 0.5 mg/ml has been investigated. The RBG is used as the output coupler. The maximum single longitude mode, Q-switched output powers of 126 mW at 692.5 kHz and 149 mW at 630.3 kHz are achieved at the pump power of 8.0 W, corresponding to the pulse durations of 144 ns and 196 ns, respectively. The single longitudinal mode output characteristics of the Q-switched laser with two-kind BP-SA solution concentrations of 1.0 and 0.5 mg/ml are compared in different optical cavity lengths. The results sufficiently validate that the multi-layered BP could be used as an optical modulator for the Q-switched single-longitudinal-mode pulse laser.

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