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PAPER

High-energy noise-like pulses generated by an erbium-doped fiber laser incorporating a PbS quantum-dot polystyrene composite film

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Abstract

We demonstrate a high-energy noise-like pulse (NLP) generated by an Er-doped fiber (EDF) laser incorporating a PbS quantum dot (QD) polystyrene (PS) composite film as a saturable absorber (SA). Benefitting from the intrinsic properties of NLPs and the high damage threshold of the PbS QD PS composite film, a stable NLP train can be obtained with an output power of 96.3 mW at a pump power of 660 mW, corresponding to a pulse energy of 9.68 nJ. This represents the highest pulse energy ever reported in an NLP EDF laser incorporating material-based SAs. The slope efficiency reached 17.37% and the steady NLP operation was able to be maintained for several hours under laboratory conditions. Our experimental results prove that the PbS QD PS composite film can serve as an effective SA to realize high-energy pulses in a mode-locked fiber laser.

1. Introduction

Recently, mode-locked fiber lasers have aroused great interest since they are capable of enabling multidisciplinary study and practical applications, such as laser spectroscopy [1], optical communication systems [2], optical sampling [3], and so on. Saturable absorbers (SAs), considered to be a critical component of mode locking, are generally classified into artificial SAs and physical SAs. Despite the fact that artificial SAs, for instance, nonlinear polarization rotation, exhibit ultrafast recovery times and support high-energy pulse generation, they are susceptible to an intracavity polarization state, making them difficult to control [4]. Nonlinear optical loop mirrors generally require long cavity lengths, leading to low fundamental repetition rates and relatively poor environmental stability [5]. Fortunately, physical SAs avoid such disadvantages and show fascinating merits, including ease of self-starting, good controllability, and excellent environmental stability, thus attracting intensive investigation [6]. So far, an enormous variety of materials have emerged to join the family of physical SAs, consisting of semiconductor SA mirrors [7, 8], graphene [9], carbon nanotubes [10], topological insulators (TIs) [11], etc. Among these, the PbS quantum dot (QD), one of the quasi zero-dimensional nanomaterials, has gained widespread attention due to its outstanding photoelectrical and mechanical characteristics [12–14]. Thanks to its large Bohr radius and size-related bandgap, its absorption peak can easily be adjusted in the range of 1–2.5 µm, which enables operational wavelength controllability [15]. In particular, the PbS QD has superior air stability, leading to the advantage of easy storage [16]. Most importantly, it has been demonstrated that PbS QDs possess a good saturation absorption effect, providing a foundation for optical pulse generation [17, 18]. Based on the above-mentioned advantages, the PbS QD is an excellent SA candidate.

In general, there are two types of PbS QD SA based on either evanescent-wave interaction or transmissive composite films. The evanescent-wave interaction type of PbS QD SA is obtained by depositing PbS QDs on the surface of a microfiber or a D-shaped fiber [19]. Although the damage threshold can be greatly improved, this type of SA is fragile and the SA properties are greatly dependent on the specialty fiber structure, which increases the complexity of the laser system. On the other hand, film-type PbS QD SAs feature good
pulses with an output power of 2.71 mW were realized [21]. The damage threshold of the PbS QD PS composite film. Q-switched pulses with a pulse energy of 586.1 nJ, providing direct evidence of the effectiveness and high portability, high flexibility, and ease of operation [20, 21]. The incorporation of this type of SA in a fiber laser can be easily realized easily by sandwiching the film between two standard fiber connectors. However, the characteristics of film-type PbS QDs are directly correlated with the polymer host. Polyvinyl alcohol, as one of the most common polymers, has been successfully used in the fabrication of PbS QD films [20, 21]. Nevertheless, it has significant disadvantages of a low glass-transition temperature and strong hygroscopicity [22], which reduce the damage threshold of the SA and undermine the benefit of easy storage of the PbS QDs. On the contrary, polystyrene (PS), another typical polymer which benefits from low thermal conductivity, high optical transmission, good heat stability, and excellent moisture resistance [23, 24] is an ideal polymer host for PbS QDs. In our previous work, a PbS QD PS composite film was successfully created and incorporated in an Er-doped fiber (EDF) laser [25]. The laser permitted the generation of high-energy Q-switched pulses with a pulse energy of 586.1 nJ, providing direct evidence of the effectiveness and high damage threshold of the PbS QD PS composite film.

In the pioneering works, most of the fiber lasers with PbS QD SAs generated Q-switched pulses [18–20, 25]. Attempts to generate mode-locked pulses using a PbS QD SA-based fiber lasers are relatively rare. In 2018, Ming et al demonstrated the first mode-locked fiber laser with a PbS QD SA, where 54 ps pulses with an output power of 2.71 mW were realized [21]. Soon afterward, pulses with a shorter pulse duration of 559 fs were obtained from an EDF laser incorporating a PbS QD SA with a high modulation depth [26]. However, the aforementioned lasers operated in the conventional soliton (CS) state, which imposed a limitation on pulse energy due to the inherent shortcomings of the CS state. Comparatively, a type of low-coherence pulse—a noise-like pulse (NLP)—supports enormous pulse energy since it is essentially a large wave packet where numerous ultrafast sub-pulses with different pulse widths and peak powers are gathered [27–30]. Therefore, the NLP fiber laser has been identified as a promising high-energy seed source.

In this report, we provide the first demonstration of a high-energy NLP EDF laser with a PbS QD PS composite film. Under an input power of 660 mW, the output power and pulse energy of the NLP train obtained are as high as 96.3 mW and 9.68 nJ, respectively. This is the highest output power and pulse energy yet reported for a real SA-based EDF laser under NLP operation. The notable signal-to-noise ratio (SNR) reaches 63.7 dB, proving outstanding stability. In addition, the laser shows a slope efficiency of 17.37% and supports stable NLP operation for several hours, indicating long-term stability. Considering the experimental results, it is believed that the PbS QD PS composite film characterized by a high damage threshold and an excellent saturation absorption effect can be considered an efficient SA to realize high-energy pulses.

2. Fabrication and characteristics of the PbS QD PS composite film

The PbS QDs used in our experiment are prepared using the improved organic metal method [16]. We employ lead chloride (PbCl₂, 99.999%, from Sigma–Aldrich) and elemental sulfur (S, 99.999%, from Sinopharm) with a Pb:S ratio of 24:1 in oleylamine (OAm, 80%–90%, from Sigma–Aldrich) as precursors. The high initial ratio of the reaction precursors contributes to a delay in the Ostwald ripening of PbS QDs, providing the possibility of synthesizing monodisperse PbS QDs. Subsequently, a purification process based on the anti-solvent method is applied in order to reduce the quantity of unreacted precursor materials. Highly monodisperse air-stable PbS QDs are then obtained. The photoluminescent (PL) emission of the PbS QDs is centered at 1610 nm with a full width half maximum (FWHM) of 60 nm, as illustrated in figure 1(a). The inset of figure 1(a) displays a photograph of the PbS QDs in a toluene solution. The particle size of the PbS QDs is approximately 7.2 nm according to the transmission electron microscopy (TEM) image depicted in figure 1(b). The uniform distribution of the PbS QDs in the toluene solution is evidenced by the narrow PL spectrum as well as the corresponding TEM image. After drying in a vacuum environment for over one day, pure PbS QD powder is mixed with PS to form a PbS QD PS composite film. The fabrication of the PbS QD PS composite film can be summarized as follows. Firstly, 20 mg PbS QD powder and 1000 mg PS powder (9.0–9.9 µm, 5% w/v, from Sigma–Aldrich) are dispersed in toluene solution, respectively. The PbS QD solution at a concentration of 40 mg ml⁻¹ and 2 ml PS toluene solution at a concentration of 500 mg ml⁻¹ are then mixed and dispersed uniformly by ultrasonication. The resulting solution is then spin-coated at 1000 rpm for 1 min on a standard glass slide to form the PbS QDs PS composite film. The absorption spectrum of the PbS QD PS composite film is shown in figure 2(a), where the absorption intensity at 1550 nm is around 0.2 and a broad absorption peak is located at 1680 nm. According to the relationship between band gap and the size of the PbS QDs [16], the size of the PbS QDs is calculated to be 7.4 nm, which coincides well with the particle size measured by TEM. Stable pulses emitted by a home-made mode-locked fiber laser with a center wavelength of 1573 nm and pulse duration of 8 ps are introduced to measure the nonlinear transmission of the PbS QD PS composite film, as shown in figure 2(b). The modulation depth, saturation fluence, and non-saturable absorption are measured at 1.17%, 0.15 µJ cm⁻², and 69.7%, respectively. The modulation depth is relatively small, but still
sufficient to be a mode locker in a fiber laser. The relatively large unsaturated loss is attributed to impurities and the aggregation of PbS QDs.

3. Laser configuration

A diagram of the mode-locked fiber laser based on the PbS QD PS composite film is schematically illustrated in figure 3. A benchtop 980 nm laser is adopted as the pump source and provides pumping light to the ring cavity via a wavelength-division multiplexer. A highly doped EDF with a length of 1 m is employed as the gain medium, whose group velocity dispersion (GVD) is \(-25.5 \text{ ps}^2 \text{ km}^{-1}\). For the purpose of maintaining unidirectional operation, a polarization-independent isolator is placed after the EDF. A 50:50 output coupler (OC) is employed to extract half the power from the cavity. The intracavity polarization can be adjusted by manipulating the settings of a variable wave retarder-based polarization controller (PC). The incorporation of the key SA is realized by directly embedding a small piece of PbS QD PS composite film with a size of \(2 \times 2 \text{ mm}\) between two fiber connectors. The rest of the cavity consists of a 19.6 m single-mode fiber with a GVD of \(-22.8 \text{ ps}^2 \text{ km}^{-1}\). The total cavity length is around 20.94 m and the net cavity dispersion is estimated to be \(-0.47 \text{ ps}^2\).

The properties of the output pulses are recorded by several measurement instruments. The pulse train is visualized by a high-speed oscilloscope (OSC, KEYSIGHT DSO90804A) with help of a 12.5 GHz high-speed biased photodetector (Newport 818-BB-51F). The radio frequency (RF) spectrum is measured by an RF spectrum analyzer (SIGLENT, SSA 3032X) while the corresponding optical spectrum is monitored by an optical spectrum analyzer (OSA, Yokogawa AQ6370C) with a 0.02 nm spectral resolution. An autocorrelator (FEMTOCHROME, FR-103XL) is used to detect the pulse autocorrelation (AC) trace.
4. Experimental results and discussion

In this experiment, continuous wave (CW) lasing emerges when the pump power is set to 59.9 mW. By increasing the pump power to 124 mW and manipulating the intracavity polarization state, NLP mode locking is initiated from the CW regime. Here, we note that the mode-locking threshold is relatively high, which may be due to the large unsaturated loss of the PbS QD PS composite film. The NLP performances are depicted in figure 4. As shown in figure 4(a), the optical spectrum of the NLPs is centered at 1568.34 nm and has a 3 dB bandwidth of 0.57 nm. Kelly sidebands do not appear in the spectrum, indicating that the laser is not working in the soliton state. The AC trace is also measured, as shown in figure 4(b). We find a double-scale structure where the pulse duration of the narrow spike is 2.36 ps assuming secant hyperbolic.
Figure 5. The NLP characteristics at a pump power of 660 mW. (a) Pulse train. Inset, a single-shot pulse of NLPs. (b) AC traces. Inset, the narrow spike of the NLPs. (c) RF spectrum. (d) Optical spectrum.

(sech²) analysis. It should be noted that the coherent spike is not centered in the measurement window, which has no effect on the measured results. The position of the coherent spike can be controlled by the delay knob of the autocorrelator. Also, there strong modulation appears around the peak, which may be caused by an imperfect adjustment of the autocorrelator. It is worth mentioning that the wave packet width cannot be measured in the AC trace, since the pedestal duration exceeds the range of our autocorrelator. Here, the single-shot pulse captured by a high-speed oscilloscope with a 25 ps temporal resolution can be used to measure the wave packet’s width as shown in the inset of figure 4(c). Specifically, many sub-pulses are contained in the wave packet and the FWHM of the wave packet reaches 0.33 ns. All of the aforementioned performances clearly indicate that the laser is operating in the NLP regime [27]. The corresponding pulse train is presented in figure 4(c). The time interval between adjacent pulses is 100.51 ns in accordance with a fundamental repetition rate of 9.949 MHz. The RF spectra of the NLPs are shown in figure 4(d). The SNR is 58.1 dB, representing excellent operating stability. The wide recorded span (1 GHz) of the RF spectrum shows that there is no Q-switching instability.

In order to achieve higher output power, we continuously increase the pump power. During this process, the laser always operates in the NLP regime. When the pump power is 660 mW, the highest output power and pulse energy of 96.3 mW and 9.68 nJ can be realized by carefully adjusting the PC. Here, we wish to stress that this is the highest output power and pulse energy recorded for the NLP EDF laser mode locked by a material SA [31–36]. We would also like to stress that the PbS QD PS composite film is not destroyed during high-power operation, indicating high damage threshold of the PbS QD PS composite film. However, the damage threshold of the PbS QD PS composite film cannot be precisely identified, owing to the limited pump power. Due to its excellent performance, it is believed that a higher output power and pulse energy could be achieved if a stronger pumping light were to be delivered. The corresponding output characteristics are illustrated in figure 5. A pulse sequence with almost identical pulse amplitudes is depicted in figure 5(a). The inset figure shows a single-shot pulse with a wave packet width of 3.06 ns, which is much wider than that of figure 4(c). The pulse duration of the narrow spike has changed to 1.60 ps (sech² assumption). The SNR of 63.7 dB provides direct evidence of good operating stability. The optical spectrum illustrated in figure 5(d) is smooth with a similar spectral profile to that of figure 4(a).
Figure 6. (a) Optical spectrum evolution of the NLP with pump powers from 160 to 610 mW. (b) Central wavelength and spectral width versus pump power. (c) Temporal evolution of the NLP with pump powers from 160 to 610 mW. (d) Spike width and wave packet width versus pump power. (e) Output power and pulse energy versus pump power.

The NLP evolutions are recorded when the pump power changes from 160 to 610 mW with a fixed intracavity polarization state, as presented in figure 6. As we can see in figure 6(a), the spectrum maintains an almost constant spectral profile with a smooth shape. The variation range of the central wavelength is only 0.15 nm, while an increase in the pump power leads to a small increase in the 3 dB bandwidth due to the increased nonlinear effect. Figure 6(c) shows the corresponding temporal evolution where the wave packet does not break up and becomes wider from 0.41 to 1.47 ns as the pump power is enhanced. This interesting behavior results from the greater quantity of the sub-pulses because of the peak power clamping effect [28]. The pulse duration of the coherent spike decreases from 2.14 to 1.64 ps, which is a good match for the 3 dB bandwidth variation of the optical spectrum. Figure 6(e) shows the output power and pulse energy along with the increase in pump power. Clearly, the output power rises from 14.78 to 93.2 mW when the pulse energy increases from 1.49 to 9.37 nJ. The slope efficiency is calculated to be 17.37% using linear regression. This is much higher than all other mode-locked PbS QD-based fiber lasers [21, 26]. The high slope efficiency is attributed to high-output-ratio OC used and the large damage threshold of the PbS QD PS composite film.

In our experiment, the laser used for the demonstration was able to operate continuously for several hours with good stability. Figure 7 shows the output power over 5 h. Clearly, except for the initial power
fluctuation, the output power of our laser remains stable. The power fluctuation is $\sim 1.4\%$. It is also noted that the PbS QD PS composite film is still effective after being stored in natural air conditions for almost two years, demonstrating excellent environmental stability. Furthermore, when the PbS QD PS composite film is taken out of the cavity, the laser cannot work in the mode-locked regime, confirming that NLP generation is due to the saturation absorption effect of the PbS QD PS composite film.

5. Conclusions

In conclusion, we report the first demonstration of high energy NLPs generated by an EDF laser incorporating a PbS QD PS composite film. At a maximum pump power of 660 mW, the noteworthy NLP output power reaches 96.3 mW. Accordingly, the pulse energy is 9.68 nJ, which is the most energetic NLP generated by a real SA-based EDF laser to date. The corresponding SNR is measured at 63.7 dB, providing direct evidence of good operating stability. The slope efficiency is estimated to be 17.37% and the long-term stability of this laser is evidenced by several hours of stable NLP operation. The experimental results not only confirm high-energy stable NLP generation but also provide direct proof of the excellent properties of the PbS QD PS composite film, such as its high damage threshold and excellent saturation absorption effect. Therefore, the PbS QD PS composite film can function as an effective SA to realize high-energy pulses in a fiber laser. Further work will identify the damage threshold of the PbS QD PS composite film and focus on applications such as supercontinuum generation and sensing using this high-energy NLP laser source. We will also try to optimize the performance of PbS QD PS for higher power and extended wavelength operation.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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