



All-Inorganic Perovskite CsPb₂Br₅ Microsheets for Photodetector Application

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Lead-halide perovskites have emerged as one kind of important optoelectronic materials with excellent performance in photovoltaic and light-emitting diode applications. Herein, we reported all-inorganic perovskite CsPb₂Br₅ microsheets prepared by a facile injection method. Through the X-ray diffraction (XRD) and Scanning Electron Microscope (SEM), it could be seen that the CsPb₂Br₅ microsheets showed single tetragonal crystalline phase and kept uniform square shape. Moreover, the as-synthesized CsPb₂Br₅ microsheets exhibited photoluminescence emission at 513 nm, and the UV–vis absorption spectrum further indicated the band gap of CsPb₂Br₅ microsheets was ≈2.50 eV. Additionally, the as-fabricated CsPb₂Br₅ microsheets based photodetector exhibited faster photoresponse characteristics of short rise time (0.71 s) and decay time (0.60 s), which demonstrated its promising application as high performance electronic and optoelectronic devices.

Keywords: perovskite, CsPb₂Br₅ microsheet, semiconductor, photoluminescence, photodetector

INTRODUCTION

Two dimensional (2D) nanostructures, such as BN [1], MoS₂ [2], and WS₂ [3] have attracted increasing attention due to the unique properties and are widely studied in many fields ranging from energy storage to environmental protection [4, 5]. Compared with one dimensional (1D) and zero dimensional (0D) nanostructures, 2D nanostructures materials show great advantages in some special applications attributed to their extraordinary electrical, optical and magnetic properties [6–8]. Recently, various kinds of semiconductor nanostructure materials are employed in photodetectors application as the reason of their high absorption coefficient, tunable bandgap, and high quantum yield [9, 10]. In the last 2 years, the halide perovskite materials were demonstrated to be amazing semiconductors with high performance. As a new family of photoelectric materials, metal halide perovskite nanocrystals have received a revival of interest based on its outstanding optoelectronic characteristics including tunable band-gap property [11], high power conversion efficiency [9], broad absorption spectrum [12], high charge carrier mobility [13], and long charge diffusion lengths [14]. However, there are few reports about the optoelectronic application based on 2D perovskite microstructure.

As one kind of the perovskites nanomaterials, all-inorganic lead halide perovskites CsPbX₃ (X = I, Br, Cl) are generally recognized as one probable substitute of organic perovskites [15, 16]. To date, all-inorganic cesium lead halide perovskite have generated considerable attention because of their higher stability and outstanding optoelectronic properties comparable to the hybrid organic–inorganic perovskites [17–20]. Thus, a large number of CsPbX₃ (X = I, Br, Cl) perovskite nanostructures such as nanocrystals [21], nanowires [22], microsheets [23, 24], nanocubes, were prepared by solution processing approach. Furthermore, the physical properties of all-inorganic nanocrystals could be adjusted by their geometric shape and size [25]. For example, Deng et al. prepared CsPbBr₃ nanocrystals with various shapes including nanocubes, nanorods, and nanoplatelets, by choosing different ligands during reprecipitation process at room temperature [26]. Therefore, more and more researchers begun to pay attention to the CsPbX₃ (X = I, Br, Cl) based photodetectors including nanofilms, nanoparticles, and nonarods. More recently, CsPb₂Br₅ as a new perovskite crystal structure has emerged as attractive semiconducting material. Wang et al. reported a new type of highly luminescent perovskite-related CsPb₂Br₅ nanoplatelets via a facile precipitation reaction [27]. Jiang's group synthesized tetragonal CsPb₂Br₅ nanosheets which was an indirect bandgap semiconductor [28]. However, there are few corresponding applications which have been further referred up to now for this kind of excellent materials. Therefore, it is interesting and necessary to carry on the study of optoelectronic application based on CsPb₂Br₅ microsheets.

In this work, we demonstrated an efficient approach for synthesis of perovskite-related uniform CsPb₂Br₅ microsheets with the size of 4.2 × 4.2 μm. The detailed structural characterization revealed that these microsheets were single-crystalline with uniform growth direction, and crystallized in pure tetragonal phase. The optical and electrical properties of the as-prepared microsheets were investigated in detail. The as-prepared CsPb₂Br₅ microsheets exhibited compositional bandgap engineering through the entire visible spectral region of 380–525 nm. PL peak appeared at 513 nm with a narrow emission line widths of 23 nm. In particular, photodetector devices based on entirely all-inorganic CsPb₂Br₅ microsheets were demonstrated for the first time. The photodetectors exhibited relatively fast rise and decay times of 0.71 and 0.60 s, respectively.

EXPERIMENTAL

The generalized protocol for synthesizing CsPb₂Br₅ perovskite microsheets was developed by modifying solution-based precipitation process initially adopted by Yang et al. [22]. Briefly, Cs₂CO₃ (100 mg), oleic acid (0.4 ml, OA), and octadecene (3.75 ml, ODE) were loaded in a 100 ml three-neck flask and heated under nitrogen flow at 120°C for 1 h to obtain Cs-oleate precursor. Then, PbBr₂ (0.36 mmol) was dissolved in ODE (5 ml) in a new 100 ml three neck flask at 120°C having nitrogen flow. After 1 h, oleylamine (0.5 ml, OLA) and OA (0.8 ml) were

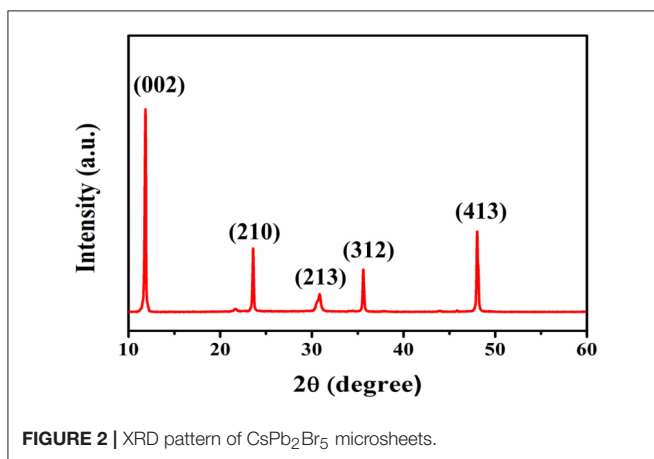
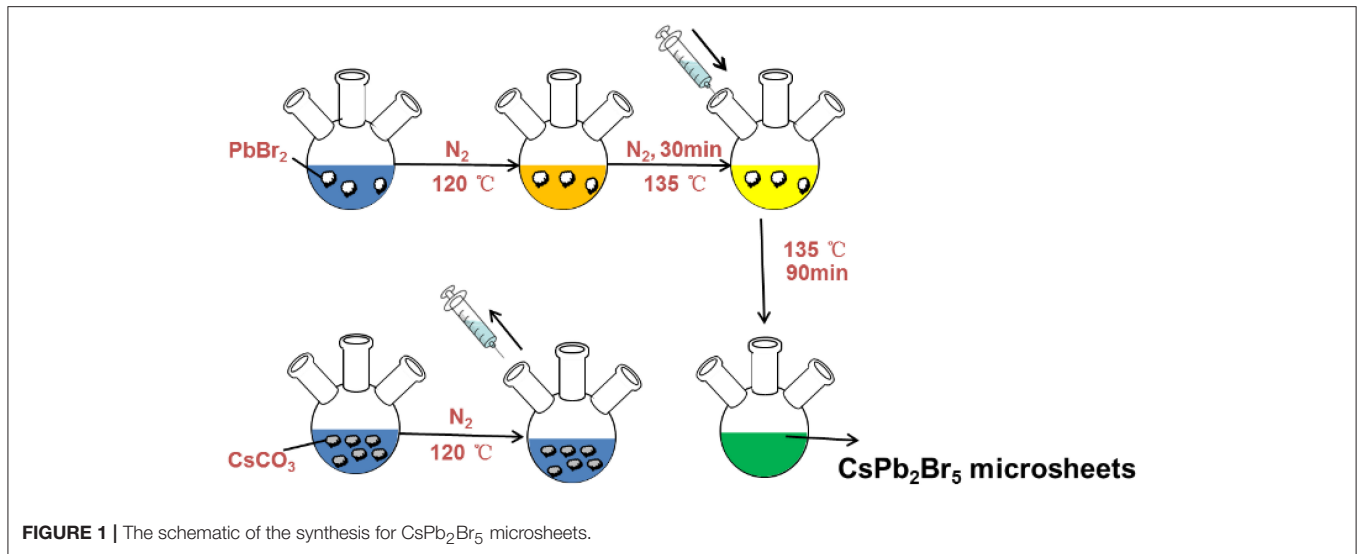
added to the mixture and heated to 135°C to keep 0.5 h, followed by swift injection of the Cs-oleate precursor (0.5 mL). The reaction was maintained with the environment of nitrogen at 135°C for 1.5 h, then, was cooled by the ice bath. The CsPb₂Br₅ product were centrifuged, precipitated, and dispersed in toluene for characterization. The schematic of synthesis for CsPb₂Br₅ microsheets was illustrated in **Figure 1**.

The crystal phases of all samples were characterized by X-ray diffraction (XRD) with Cu Kα radiation (XRD-6100, SHIMADZU, Japan). The surface morphology and composition were observed by scanning electron microscopy (SEM, JSM-7800F) with X-ray energy dispersive spectrometry (XEDS). Atomic force microscopy (AFM) imaging was carried out on a scanning probe microscope (Nanonavi, SPA-400SPM, Japan) using a tapping mode. The absorption spectra was adopted by a Scan UV-vis spectrophotometer (UV-vis: UV-2100, Shimadzu, Japan), while photoluminescence (PL) spectra were measured by a fluorescence spectrophotometer (PL: Agilent Cary Eclipse, Australia) which included a Xe lamp as an excitation source with optical filters). The transmission electron microscopy (TEM) and high-resolution TEM (HRTEM) images were obtained using a ZEISS LIBRA 200FE microscope. The on/off photocurrent ratio of the CsPb₂Br₅ microsheets was obtained by a source meter (Keithley 4200).

RESULTS AND DISCUSSION

To get clear information about the CsPb₂Br₅ microsheets, the powder X-ray diffraction (XRD) was used to characterize the crystallographic structure of the as-obtained CsPb₂Br₅ microsheets. **Figure 2** shows XRD pattern of the CsPb₂Br₅ microsheets, all of the characteristic diffraction peaks could be indexed into a tetragonal phase (PDF#25-0211), which was in good agreement with literature data for the tetragonal perovskite structure [27]. The crystal planes were marked on the XRD pattern. The diffraction peaks were strong and sharp, which indicated that the obtained CsPb₂Br₅ microsheets were highly crystalline. Moreover, there were no any impurity peaks detected in the sample, suggesting its high crystalline quality of the as-prepared CsPb₂Br₅ microsheets.

Additionally, in order to study the morphology of CsPb₂Br₅ microsheets, Scanning Electron Microscope (SEM) was employed for observation. From **Figure 3a**, it could be seen that the as-synthesized CsPb₂Br₅ microstructures were square shape with average lateral size (4.2 × 4.2 μm), and there were few by-products, which suggested the high purity of CsPb₂Br₅ microsheets. X-ray energy dispersive spectrometry (XEDS) measurement was performed to identify the composition of the as-obtained microsheets, as shown in **Figure 3b**. It could be seen that the composition elements were determined as Cs, Pb, and Br elements, and the Cs/Pb/Br atomic ratio was determined as 11.7/22.5/65.8. To investigate the distribution states, XEDS elemental mappings were carried out on the surface of CsPb₂Br₅ microsheet. **Figure 3c** shows the single typically CsPb₂Br₅ microsheet, and accordingly elemental XEDS mappings (**Figures 3d–f**) were measured in this area. The XEDS



elemental mapping images further indicated the homogeneous distribution of Br (**Figure 3d**), Cs (**Figure 3e**) and Pb (**Figure 3f**) elements within individual microsheet. The thickness of as-synthesized CsPb₂Br₅ microsheets was measured by atomic force microscope (AFM). The AFM image (**Figure 3g**) and line profile (**Figure 3h**) showed that the thickness of the CsPb₂Br₅ microsheets was about 21 nm.

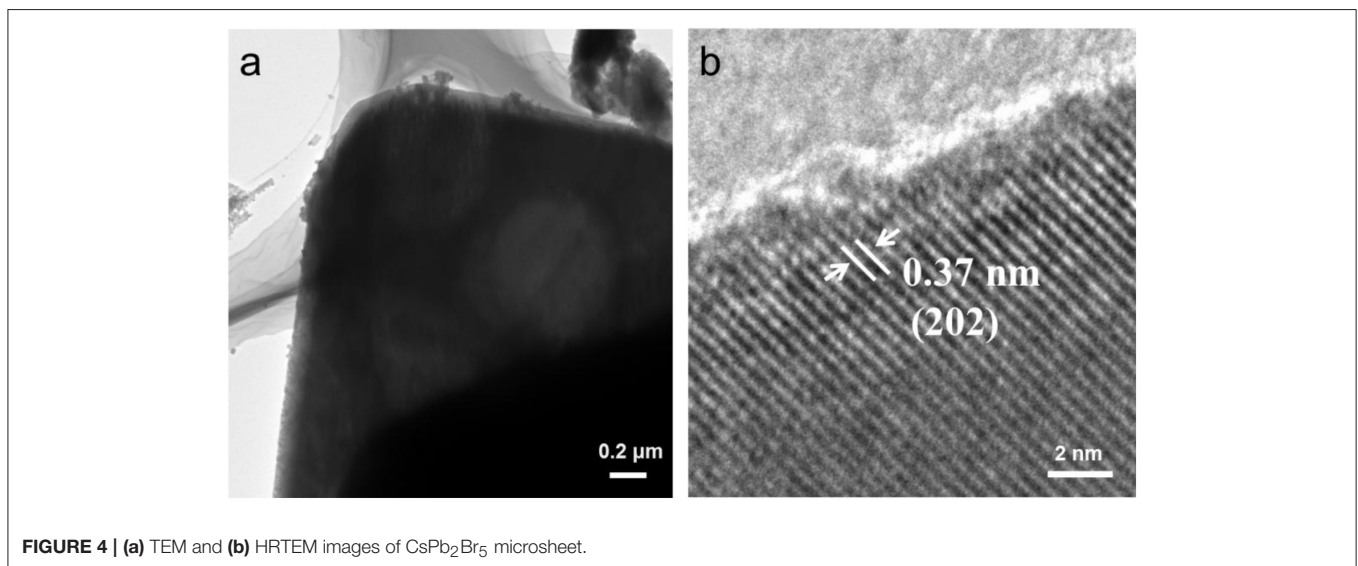
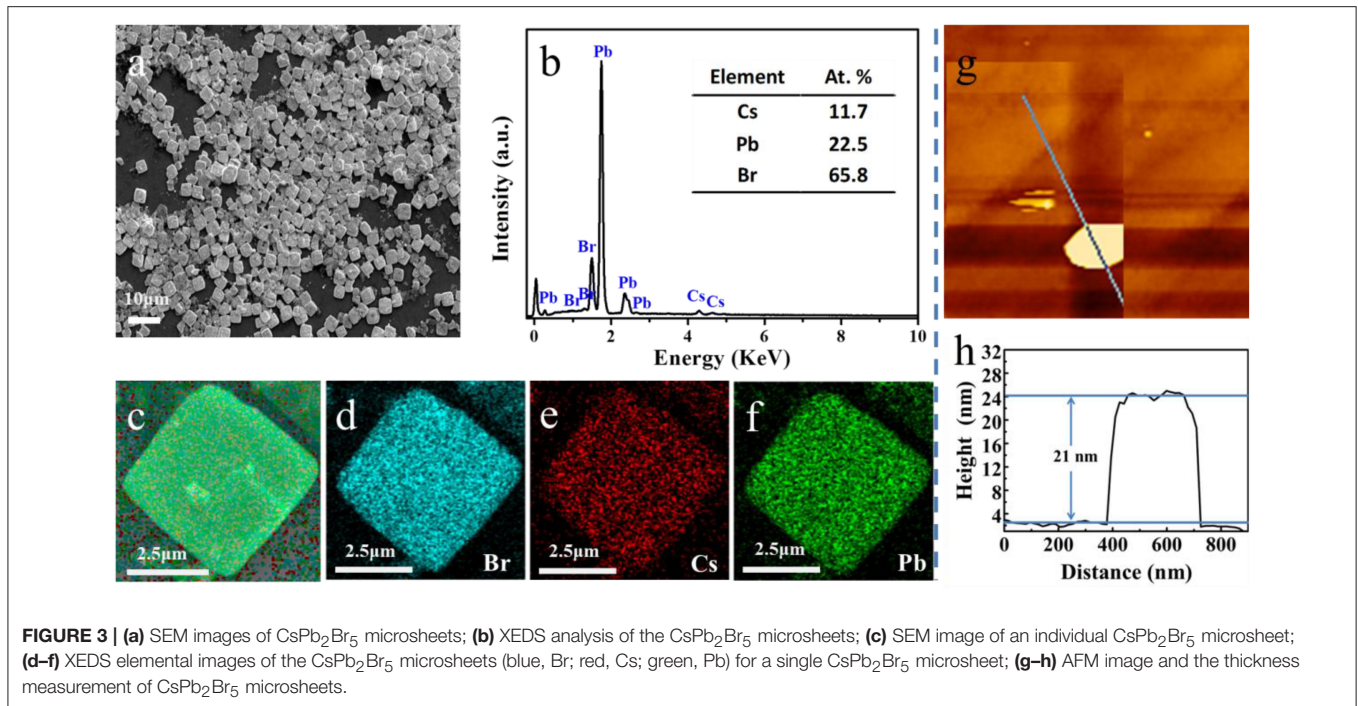
The morphology of the CsPb₂Br₅ thin microsheet (4.2 × 4.2 μm) was further tested by typical TEM, as showed in **Figure 4a**. **Figure 4b** was the high-resolution transmission electron microscopy (HRTEM) image of single CsPb₂Br₅ microsheet, it could be obviously observed that the interplanar distances was about 0.37 nm, which could be assigned as the lattice (202) planes of the tetragonal structure. And the clear lattice also confirmed the CsPb₂Br₅ microsheets had high quality crystalline, which matched well with the XRD results.

As the excellent properties of the as-prepared CsPb₂Br₅ microsheets, it has been studied as lasing application in our previous work [29]. Herein, the large lateral dimensions of these perovskite microsheets motivated us to explore their potential

applications in optoelectronic devices. As shown in **Figure 5a**, a simple photodetector device was fabricated by dropping the CsPb₂Br₅ microsheets onto gold interdigital electrode with 3 μm spacing between adjacent fingers. The light source used in this device was a continuous wave laser (excitation at 405 nm with an optical power of 20 mW), otherwise, the bias voltage could be adjusted from 1 to 30 V for testing the photoresponse activity. In order to clearly illustrate the structure of the device, a typical SEM image of the CsPb₂Br₅ microsheets based photodetector is showed in **Figure 5b**. It could be observed that some of the as-prepared CsPb₂Br₅ microsheets were successfully crossed on two gold electrodes, which demonstrated the good devices.

The optical properties of the CsPb₂Br₅ microsheets films were characterized by UV-vis absorption and photoluminescence (PL) spectra, as showed in **Figure 6A**. The CsPb₂Br₅ microsheets had an absorption spectrum that was dominated by sharp exciton peaks, as similar to the optical features of previously reports [24, 27]. Furthermore, the absorption intensity was lower than the orthorhombic CsPbBr₃, further suggesting that the CsPb₂Br₅ microsheets were successfully obtained [28]. The absorption spectrum (blue line) exhibited an absorption peak at around 495 nm, yielding an excitonic bandgap of about 2.50 eV [30, 31]. The PL emission spectrum (red line) exhibited a highly symmetric form located at 513 nm (≈2.42 eV) with a narrow full width at half maximum (FWHM) of 23 nm. No sub-bandgap emission was observed in the PL spectrum, indicating that CsPb₂Br₅ microsheets can be employed in photodetectors [32]. PL properties of the CsPb₂Br₅ were convinced by our group, and the CsPb₂Br₅ can remain stable under ambient environment [29, 33].

Current-voltage (I-V) characteristics of the CsPb₂Br₅ microsheets based photodetector under the dark and illumination with 405 nm light are illustrated in **Figure 6B**. The room temperature I-V curves were measured at different bias voltage ranging from -8 to 8 V in air. Clearly, I-V curves presented linear dependence on the applied bias, indicating a good ohmic contact between CsPb₂Br₅ microsheets and gold



electrodes [34]. The photo-excited current increased by more than 13 times compared with the dark current, indicating the ultimately high sensitivity of the photodetector. The increase in current under illumination could be attributed to the large amounts of electron-hole pairs generated by the photon absorption and subsequently extracted by the electrical field [35].

Photoresponsivity is also one critical factor used to evaluate the performance of photodetectors. **Figure 6C** illustrates the photoresponse behavior of the photodetector based CsPb₂Br₅ microsheets, which was measured in the dark and with 405 nm

illuminating periodically at a different bias of 8, 10, 20, and 30 V, respectively. It could be observed that upon illumination, the photocurrent rapidly increased drastically due to the increase in carrier drift velocity and then drastically decreased to its initial level when the light was turned off, indicating the higher stability and reproducible characteristics of the photodetector device [35]. Also, it could be seen that the photocurrent increased when the applied voltage was elevated. At a bias of 30 V, the dark current was 0.03 μA and when the flexible device was under illuminated, the photocurrent increased to 0.89 μA, showing a photocurrent on/off ratio of 30. It should be noted that the applied bias voltage

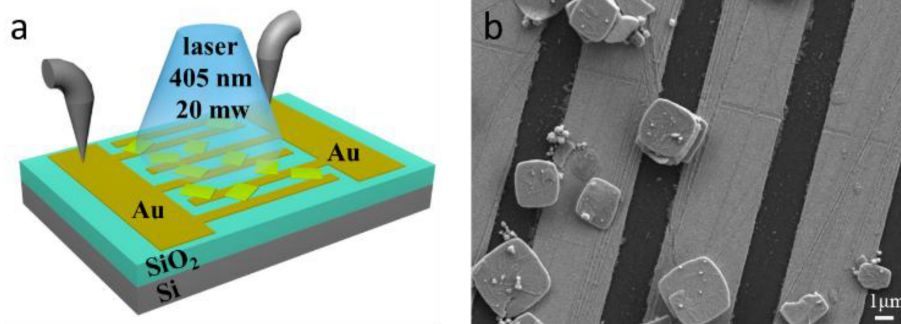


FIGURE 5 | (a) Schematic of a photodetector device based on CsPb₂Br₅ microsheets. (b) The SEM image of real photodetector device.

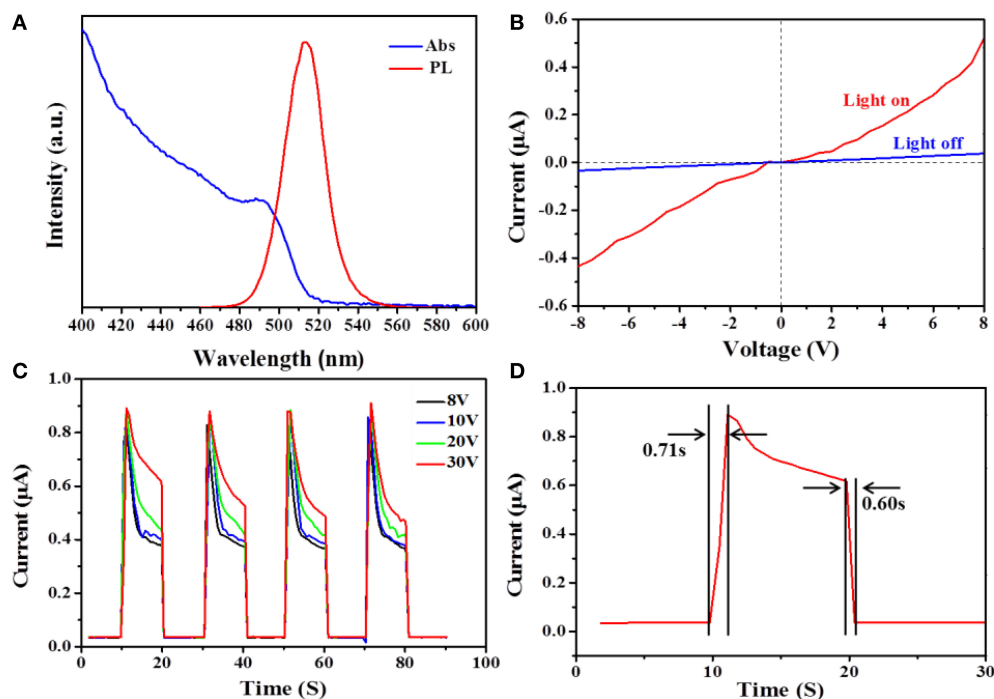


FIGURE 6 | (A) Spectrum of fluorescence (excited by light with $\lambda = 365$ nm) (red line) and absorption (blue line) for CsPb₂Br₅ microsheets. (B) I–V curves of the photodetector measured in the dark and under illumination using a 405 nm laser diode by sweeping the voltage from -8 to 8 V. (C) Photocurrent–time response of the photodetector measured in the dark and with 405 nm illuminating with a bias of 8 , 10 , 20 , and 30 V. (D) The rise time and the decay time of the photo-detector device.

influenced the on/off ratio of the devices, which was caused by the exciton dissociation and the background current [36].

The time response speed is usually recognized as one key factor for evaluating the performance of sensor and it could determine the capability of photodetector. **Figure 6D** shows the response time and recovery time of our device, which were found to be around 0.71 and 0.60 s, respectively. Both of them are shorter than 1 s, which are significantly faster compared with the previously reported perovskite detectors [37–39]. And, the faster response speed of this CsPb₂Br₅ thin microsheets based photodetector could be ascribed to the high crystal quality of as-prepared CsPb₂Br₅ microsheets, which guaranteeing the efficient

optical absorption and photogeneration of carriers. On the other side, the short transit time and large surface-to-volume ratio of CsPb₂Br₅ thin microsheets tend to induce defects and dangling bonds on the surface of microsheets [40, 41]. The switching in the two states exhibited faster photoresponse characteristics, allowing the device to act as a high-quality photosensitive switch.

CONCLUSIONS

In summary, we have synthesized the CsPb₂Br₅ microsheets through a low-cost injection method. The characterization results

of XRD, SEM, and HRTEM confirmed that the as-grown CsPb₂Br₅ microsheets were single crystalline and had uniform tetragonal morphology. The optical band gap of the CsPb₂Br₅ microsheets was found to be ≈ 2.50 eV and the PL emission peak was located at around 513 nm with a 23 nm FWHM. Besides, photodetector based on CsPb₂Br₅ microsheets was fabricated and studied for the first time, exhibiting great photoresponse with the response time (0.71 s) and decay time (0.60 s). All these unique characteristics suggested that CsPb₂Br₅ microsheet is a promising material for photodetection applications.

AUTHOR CONTRIBUTIONS

XT did the major work of this manuscript including synthesis process, characterization, and writing; SH, ZZu, ZH, and SL

synthesized part of perovskite CsPb₂Br₅ microsheets; WH, DZ, and ZZa gave some supports on the TEM and SEM testing; JD gave some suggestion and comments on writing paper.

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Conflict of Interest Statement: 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.

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