

Research article

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Electroluminescence and photo-response of inorganic halide perovskite bi-functional diodes

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Abstract: In this work, we report our efforts to develop a novel inorganic halide perovskite-based bi-functional light-emitting and photo-detecting diode. The bi-functional diode is capable of emitting a uniform green light, with a peak wavelength of 520 nm, at a forward bias of >2 V, achieving a high luminance of $>10^3$ cd/m² at 7 V. It becomes an efficient photodetector when the bi-functional diode is operated at a reverse bias, exhibiting sensitivity over a broadband wavelength range from ultra-violet to visible light. The bi-functional diode possesses very fast transient electroluminescence (EL) and photo-response characteristics, e.g. with a short EL rising time of ~ 6 μ S and a photo-response time of ~ 150 μ S. In addition, the bi-functional diode also is sensitive to 520 nm, the wavelength of its peak EL emission. The ability of the bi-functional diodes for application in high speed visible light communication was analyzed and demonstrated using two identical bi-functional diodes, one performed as the signal generator and the other acted as a signal receiver. The dual functions of light emission and light detection capability, enabled by bi-functional diodes, are

very attractive for different applications in under water communication and visible light telecommunications.

Keywords: halide perovskite; light-emitting diode; photo-detector; visible light communication.

1 Introduction

Bi-functional devices enabling light emission and photo detection in one configuration are attractive for various applications and valuable for simplifying fabrication processes and reducing the device size [1–5]. To obtain such bi-functionality, one strategy is to stack a photodiode (PD) and a light emitting diode (LED) together with an inter electrode between the two units [1, 2]. This includes a tedious control of the inter electrode and precise design of the multi-layer configuration. Another method is to employ a photo sensitive layer in the LEDs [3–5], where the energy levels of the interlayers should be well aligned and most of the materials are not solution processable.

In the past years, great progress has been made in both organic emitting materials and their light emitting diodes (OLEDs), but the bigger full width at half maximum (FWHM) for OLEDs make them unsuitable applied in photo detection field [6–8]. By contrast, perovskite materials are emerging as promising candidates in a variety of applications due to their excellent optoelectronic properties, such as high carrier mobility, large absorption coefficient and saturated light emission characteristics [9–16] for various applications in solar cells [9–12] and light emitting devices [13–16]. The inorganic halide perovskite, e.g. cesium lead bromide (CsPbBr₃), is known for its good thermal stability and excellent optoelectronic properties [16–24]. The CsPbBr₃ perovskite is intensively investigated for application in both solar cells [17–19] and LEDs [15, 20, 21] due to its simultaneously excellent light absorption and saturated light emission properties. In CsPbBr₃ based solar cells, efficiency up to 6.7% was achieved indicating CsPbBr₃ as an excellent light sensitive material [19]. Meanwhile, CsPbBr₃ also is suitable for photo detection applications with high sensitivity and responsivity [22–24]. On

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the other hand, with an exceptional photo luminance quantum yield (PLQY), CsPbBr₃ perovskite draws substantial attentions for application in LEDs with unique emission characteristics, e.g. narrow emission spectrum and low turn-on voltage [20, 21]. CsPbBr₃ perovskite with simultaneous excellent light harvesting and light emitting properties make it a very suitable candidate for use in multi-functional optoelectronics.

In this work, we report our efforts to develop high performing CsPbBr₃ perovskite-based bi-functional diodes. The CsPbBr₃ perovskite functional layer in the bi-functional diode serves as a light emission layer when the diode is operated at a forward bias, and it becomes a light detection layer while the diode is operated under a reverse bias. The excellent optoelectronic properties of the CsPbBr₃ perovskite layer, such as saturated green light emission and good light absorption in the ultraviolet (UV)-visible light wavelength range, enable the realization of the electroluminescence (EL) and photo-response dual function in one device. The EL characteristics, functioning as a LED, and photo-response properties, acting as a PD, of the bi-functional diode can be realized by controlling the polarity of the external biases. The unique EL and photo-response dual-function in one identical device has the advantage in reducing the complexity of device configuration, which often involves the use of tandem structure, and thereby simplifying the fabrication procedures. It was also found that the bi-functional diodes exhibit very fast transient EL and transient photo-response characteristics, which are a prerequisite for application in optical communication. A visible light communication system operated at 520 nm was demonstrated utilizing two identical bi-functional diodes, one functioned as the signal generator and the other acted as the signal receiver. The outcomes of the work are very encouraging. It is anticipated that the inorganic halide perovskite bi-functional diodes can be a commercial viable technology for applications in, e.g. Internet of Things, smart lighting, indoor visible light and under-water communication etc.

2 Methods

The CsPbBr₃ precursor was prepared in a non-stoichiometric manner with excess CsBr [9, 20]. 0.18 M PbBr₂ (Sigma Aldrich) and 0.324 M CsBr (Sigma Aldrich) were dissolved in dimethyl sulfoxide (Sigma Aldrich) and kept stirring before use. All chemicals are used as received. 180 nm thick pre-patterned indium tin oxide (ITO)-coated glass substrates, with a sheet resistance of 100 ohm per

square, were cleaned by ultra-sonication sequentially with diluted detergent, deionized water, acetone and isopropyl alcohol each for 15 min. After drying in oven, the ITO/glass substrates were exposed to the UV-Ozone for 10 min to remove the residual chemicals on ITO surface. The UV-Ozone treatment also helps to improve the surface hydrophilicity assisting in the deposition of the Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) hole transporting layer (HTL) in air. A 40 nm thick PEDOT:PSS HTL was optimized by controlling the spin coating conditions. The substrates with PEDOT:PSS thin film were annealed at 140°C for 10 min to remove the residual water and then transferred into glove box with oxygen and H₂O levels below 0.1 ppm. Afterwards, the CsPbBr₃ thin film was grown on the surface of the PEDOT:PSS HTL using two different approaches, e.g. (1) drop-spin method and (2) spin-drop method. In the drop-spin method, 80 μl precursor was dropped to the surface of PEDOT:PSS HTL and the substrate rotation was started only after the perovskite precursor solution was spread evenly across the substrate. While in the spin-drop method, substrate rotation occurred prior to the drop of the 80 μl precursor solution. The thickness of the CsPbBr₃ layers was controlled by the rotation speed in the spin-coating process, e.g. the CsPbBr₃ thicknesses of 26 nm, 30 nm and 36 nm were prepared using different rotation speeds of 3500 rpm, 3000 rpm, and 2500 rpm for 60 s, respectively. The CsPbBr₃ thin films were annealed at 70°C for 10 min in the glove box, the samples were transferred to the vacuum chamber for deposition of 100 nm thick 2,2',2''-(1,3,5-Benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) electron transportation layer (ETL), 1 nm LiF electron injection layer (EIL) and 100 nm thick Al electrode, prepared by thermal evaporation. The devices were encapsulated in the glove box before the measurements. The active area of the device is ~0.1 cm².

The UV-visible absorption spectra of the CsPbBr₃ active layers were recorded using a HP 8453 spectrophotometer. The morphological characteristics of the thin film were investigated using scanning electron microscope (SEM) by a LEO 1530 system. The crystallinity of the CsPbBr₃ active layer was measured by X-ray Diffraction (XRD) technique. The photoluminescence (PL) properties of the thin film were characterized using a He-Cd laser operated at 325 nm as the activation light. The luminance–current density–voltage (L – J – V) characteristics of the bi-functional diodes were characterized using a luminance colorimeter, an electro meter and control software. Two LED (Zolix) light sources with different peak EL wavelengths of 365 nm and 450 nm were used for measuring the photo-response behavior of the bi-functional diodes. The transient EL of

the bi-functional diodes was detected using a commercial Si photodiode (Thorlabs) and recorded by an oscilloscope (Tektronix). The transient photo-response of the PDs was recorded on an oscilloscope under pulsed blue light (450 nm). All the optoelectronic measurements were conducted in ambient.

3 Results and discussion

In inorganic halide perovskite CsPbBr₃-based optoelectronic devices, huge attention has been devoted to the synthesis of CsPbBr₃ quantum dots (QDs) [21, 22, 25–27] for high performance, e.g. PLQY [27, 28]. However, the synthesis procedures usually include complex passivation of the QD surfaces which causes injection barriers due to the surface ligands [29]. Thin film CsPbBr₃ perovskite based devices are becoming a promising choice for functional optoelectronics [30–33] due to the ease of fabrication using costless techniques like spin coating and spray coating. The morphology of the CsPbBr₃ emission layer plays a critical role in achieving high luminous efficiency of CsPbBr₃-based LEDs [34, 35].

In this work, it is found that the coating method of the CsPbBr₃ layer has a huge impact on the thin film morphology. The intrinsic fast crystallization of the CsPbBr₃ perovskite tends to form different morphological features depending on the spin coating methods. In the drop-spin method where the substrate rotation was set after the casting of the precursor, the fast crystallization of CsPbBr₃ tends to form ring-like features during the slow free spread as schematically shown in Figure 1A. The left insert in Figure 1A is a photo taken for CsPbBr₃ thin film on ITO/HTL with clear ring like features under 350 nm UV light. The LEDs based on these ring-featured thin films also exhibit un-uniform light emission as shown in the right insert in Figure 1A. The clear ring features observable to the naked eye in the CsPbBr₃ thin film is attributed to the fast crystallization of the CsPbBr₃ material during the slow precursor spread. The ring-like feature of the film is formed due to the distribution of the spatially varying layer thickness. The corresponding profile of the layer thickness across the CsPbBr₃ surface is clearly seen in the step profiler measurement. Figure S1 (in Supporting Information) shows the photograph taken for the measurement using the step-profiler and the direction of the line scanning in the measurement. The line profile of the ring-like surface was given in Figure S2. Clear height peaks can be seen in the profile of the film thickness along the scan direction, corresponding to the position of the “ring” feature in the film. The schematic diagram illustrating the

formation of the ring-like features is shown in Figure S3. The precursor was casted on the substrate and spread freely in the film formation. During this period, the ring-like features were formed due to the fast crystallization of the CsPbBr₃ film. To deal with this inhomogeneous thin film formation, a spin-drop method was introduced where the precursor was casted on a rotating substrate. This method provides a centrifugal force during the precursor spread as schematically illustrated in Figure 1B. As a consequence, a highly repeatable uniform CsPbBr₃ thin film was obtained as the photograph shown in left insert of Figure 1B under UV light. The uniform surface profile of the CsPbBr₃ thin film is presented in Figure S4, without any height peaks that were seen in the ring-like films. The LED with the uniform CsPbBr₃ thin film shows homogeneous light emission as the photo shown in right insert of Figure 1B. The centrifugal force provided by the rotating substrate assists the precursor spread which is responsible for the formation of highly reproducible and uniform CsPbBr₃ emission layer.

The characteristics of the uniform CsPbBr₃ thin films were investigated by SEM and XRD techniques. The top view SEM image of CsPbBr₃ thin film is shown in Figure 2A. Uniformly distributed CsPbBr₃ crystals with relative small sized polycrystalline features were obtained [20, 36]. The spinodal decomposition mechanism is likely to play a role in the formation of the CsPbBr₃ thin film, leading to a finely dispersed microstructure, as shown in Figure 2A. The uniformly distributed small CsPbBr₃ crystal grains have the advantages for light emission. The XRD pattern shown in Figure 2B, exhibits three typical diffraction peaks at 15.2, 21.5, and 30.4 degree, which could be assigned to the (100), (110) and (220) planes of the CsPbBr₃ crystal, identifying the formation of

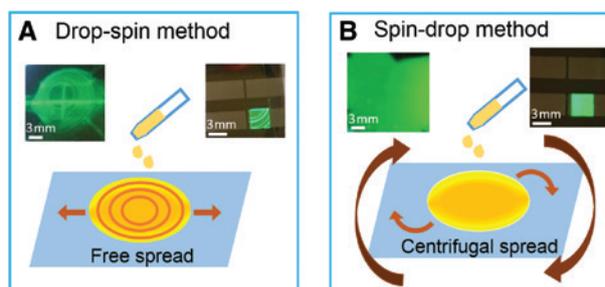


Figure 1: Schematic diagrams illustrating the quality of the CsPbBr₃ thin films prepared by (A) drop-spin and (B) spin-drop methods. The inserts in (A): the photo pictures taken for the CsPbBr₃ thin film grown on PEDOT:PSS/ITO exposed under UV light (left) and a bi-functional diode emitting light at 3 V (right). The inserts in (B): the photo pictures taken for the CsPbBr₃ thin film grown on PEDOT:PSS/ITO exposed under UV light (left) and a bi-functional diode emitting a uniform light at 3 V (right).

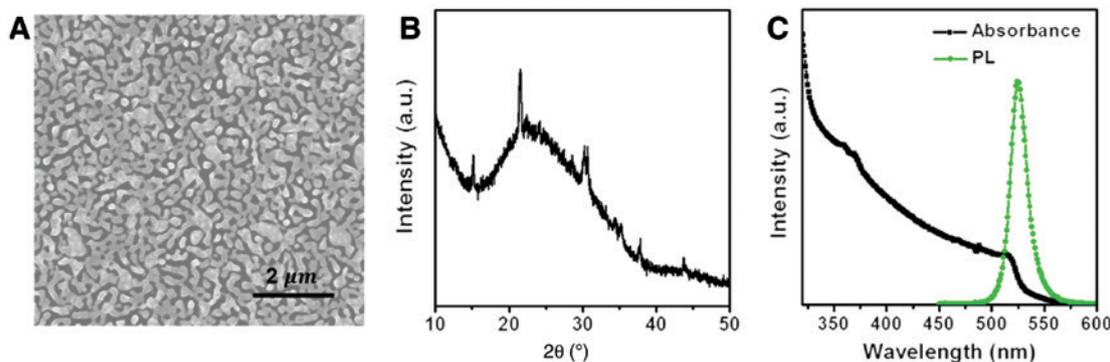


Figure 2: CsPbBr₃ perovskite thin film characteristics.

(A) Top view SEM image, and (B) XRD spectrum measured for the CsPbBr₃ perovskite thin film deposited on the PEDOT:PSS /ITO substrates, and (C) absorption and PL spectra measured for the CsPbBr₃ perovskite thin film deposited on the quartz substrates.

orthorhombic structure of the crystal [17, 33]. The absorbance and static PL spectra of CsPbBr₃ thin film are illustrated in Figure 2C. Clearly, the maxima of the PL signal is located near the absorption edge with an insignificant difference indicating the PL emission of CsPbBr₃ originates from direct recombination of charges across the band edge of CsPbBr₃ [20]. The absorption spectrum shows CsPbBr₃ thin film is capable of absorbing photons with energy higher than its bandgap of ~2.3 eV [19, 36], such that it is a good light absorber for UV and portion of visible light. The PL spectrum of CsPbBr₃ thin film is very narrow with a FWHM of ~20 nm, indicating CsPbBr₃ perovskite is an appealing material for saturated light emission. These unique properties featuring simultaneously excellent light absorption and emission suggest that CsPbBr₃ perovskite is a versatile material for multi-functional optoelectronic devices.

The diode consisting a CsPbBr₃ thin film as the active layer with a configuration of ITO/PEDOT:PSS/CsPbBr₃/

TPBi/LiF/Al was fabricated and characterized. The CsPbBr₃ layer was sandwiched between a HTL PEDOT:PSS and an ETL TPBi as schematically shown in Figure 3A, which is a typical structure in CsPbBr₃ perovskite based LEDs [30, 37, 38]. The diode possesses bi-functionality by controlling the bias polarity as depicted in Figure 3A. With forward bias, the diode works in the LED mode where holes and electrons are injected to the CsPbBr₃ emissive layer as shown in Figure 3B, resulting in EL. While with reverse bias, the diode function as a PD in which photo generated holes and electrons are formed and then collected by respective electrodes driven by the external electric field. The schematic working mechanism is shown in Figure 3C. This EL and photo-response bi-functionality in one device takes advantage of the versatile properties of the CsPbBr₃ perovskite active layer.

The EL properties of the bi-functional diodes were firstly investigated and the performances were optimized

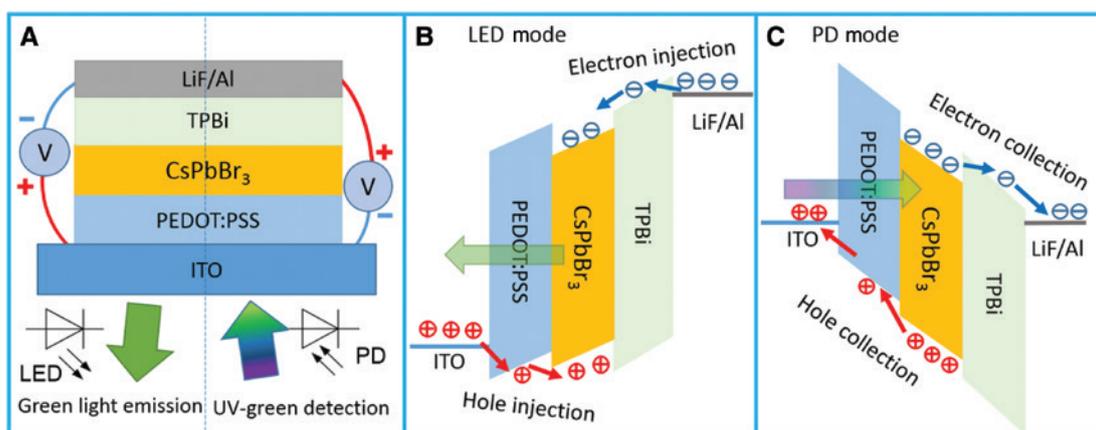


Figure 3: Schematics of the working mechanism of the bi-functional diodes.

(A) Schematic cross-sectional view of the bi-functional diode and its EL and photo-response dual-function. Schematic diagrams illustrating (B) charge injection for light-emitting process when the bi-functional diode is operated at a forward bias and (C) charge extraction for light detection when it is operated at a reverse bias.

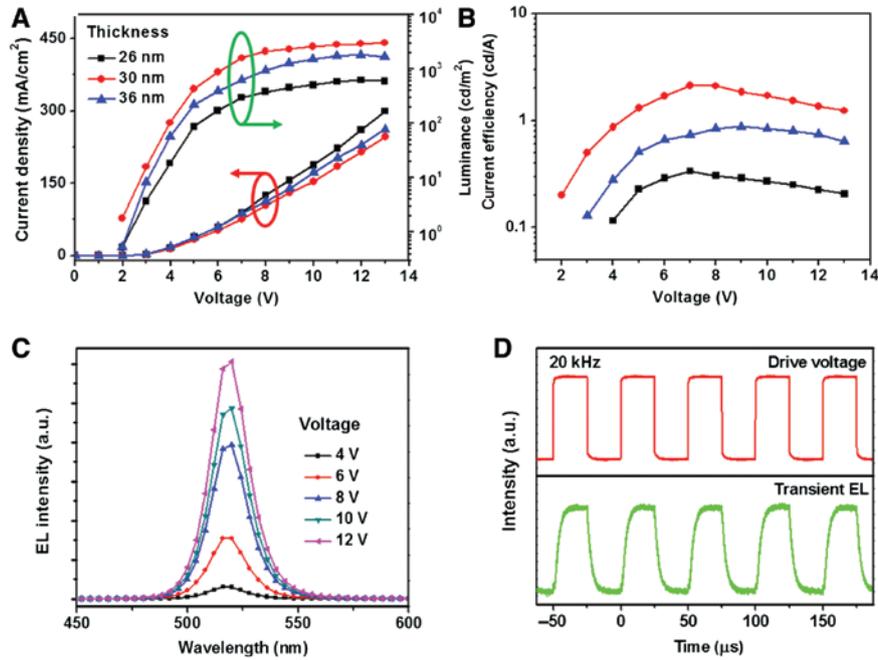


Figure 4: Electroluminescence characteristics of the bi-functional diodes.

(A) J - L - V characteristics and (B) current efficiency measured for the bi-functional diodes with different active layer thicknesses of 26 nm, 30 nm and 36 nm. (C) Typical EL spectra measured for the bi-functional diode at different forward biases. (D) The transient EL characteristics of the bi-functional diode operated by a 5 V 20 kHz-modulated power source. The profile of the 20 kHz-modulated bias also is plotted for comparison.

by controlling the thickness of CsPbBr_3 perovskite layer. The L - J - V characteristics were shown in Figure 4A. With increasing the forward bias, the current density as well as the luminance of all LEDs increases. And all three devices with different active layer thickness exhibit a low turn-on voltage of ~ 2 V. The ultra-low turn-on voltage even lower than the band gap of the material (~ 2.3 eV) suggests an extremely efficient charge injection from charge transporting layers to the active layer [36]. The 30 nm-thick CsPbBr_3 perovskite active layer based LED shows relatively higher luminescence as compared to the other two devices with the thickness (26 and 36 nm) of CsPbBr_3 layer, achieving a maximum luminance of 3035 cd/cm^2 at a forward bias of 13 V. The current efficiency of the three LEDs as a function of active layer thickness and bias voltage are given in Figure 4B. The LED with a thickness of 30 nm for active layer achieved higher current efficiency due to an enhanced EL performance. All LEDs exhibit a voltage independent emission spectrum.

The EL spectra of the LED with 30 nm-thick CsPbBr_3 perovskite layer were plotted in Figure 4C as a function of bias voltage. Identical EL peak were observed regardless the bias voltage. The EL spectra were also in good agreement with the PL characteristics. The transient EL performance of the diodes in the LED mode was also investigated. A function generator was employed serving as the power supply. The transient EL of the diode was detected

using a commercial Si photodiode and recorded by oscilloscope. In Figure 4D, the transient EL were compared to the corresponding driving signal of the functional generator at a frequency of 20 kHz. The rise and fall time of EL was measured to be $\sim 6 \mu\text{s}$ suggesting the LEDs are potentially suitable for transient light generators in high speed visible communication systems.

The PD characteristics of the bi-functional diode were then carried out. The CsPbBr_3 perovskite active layer was controlled to be 30 nm. In Figure 5A, the external quantum efficiency (EQE) and photoresponsivity (R) of the bi-functional diode as a function of wavelength operated at 0 V were presented. The photoresponsivity defined as $R = I_{\text{photo}}/P_{\text{in}}$, where I_{photo} is the photocurrent and P_{in} is the incident light power, has a close relationship with EQE in the form of $R = EQE/h\nu$, where R is in A/W and $h\nu$ is the incident photon energy in eV. These results indicate the diode can be operated in a self-powered mode where photo-generated charges could be collected under the built-in potential. The EQE of 7% and photoresponsivity of 20 mA/W were obtained at 350 nm. The collection efficiency of the photo-generated charges can be largely improved by applying a reverse bias on the PD. The photo-response of the bi-functional diodes under reverse bias was measured under light illumination of 365 nm and 450 nm. The current-voltage characteristics of the diodes under 365 nm and 450 nm light illumination were presented in Figure 5B. The bi-functional diode shows

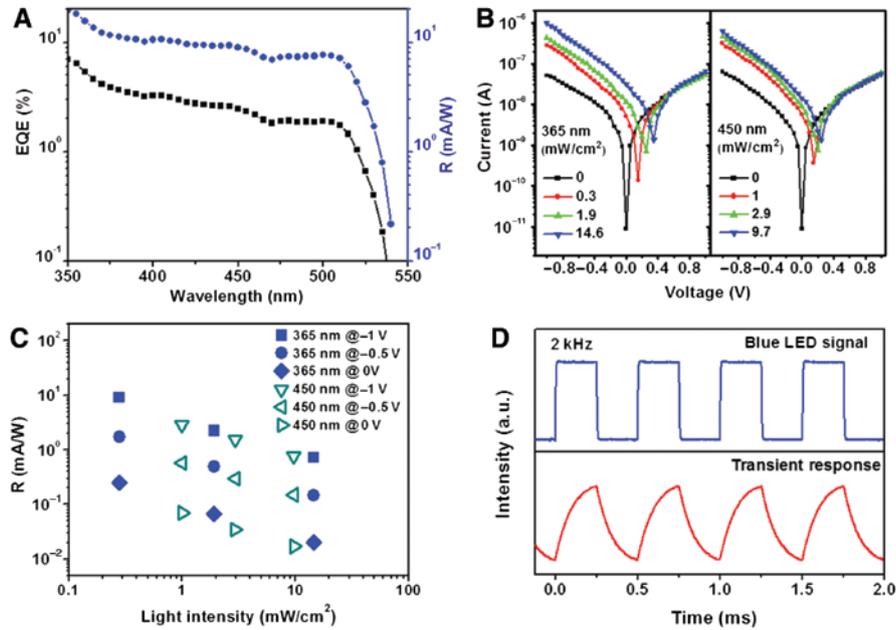


Figure 5: Photo-response properties of the bi-functional diodes.

(A) EQE and corresponding photoresponsivity of the bi-functional diode at 0 V bias, (B) the photo current–voltage characteristics measured for the bi-functional diodes using two LED sources with different peak EL wavelengths of 365 and 450 nm under different light intensities. (C) The photoresponsivity–light intensity characteristics measured for the bi-functional diode operated at different reverse biases using two LED light sources. (D) The transient photo-response of the bi-functional diode operated at 0 V under a 2 kHz-modulated 450 nm blue light LED source.

good photo-response both under 365 nm and 450 nm. With the increase of the reverse bias, the photo current increases due to enhanced charge collection with the help of external electrical field. In Figure 5C, the extracted photoresponsivity of the diodes as a function of reverse bias and light intensity were summarized. The photoresponsivity of the diodes show a light intensity dependent manner similar to that of the commercialized photodetectors. At a higher reverse bias, the photoresponsivity enhances due to improved collection efficiency of the photo-generated charges. Compared to the results of the CsPbBr₃-based photodetectors [20, 21], the devices benefit from a lower driving voltage which is potentially attractive for low-power portable and wearable applications. The transient photo-response of the bi-functional diode was estimated by applying a series of blue light signals to the device and recording the photo current signals. The profile of the blue light signals and the photo-response of the diode operated at 0 V are given in Figure 5D. According to the transient photo-response profile, the rise and fall time of the diode were estimated to be ~160 and ~170 μ S. The transient photo-response was fast as compared to that of the reported CsPbBr₃ based detectors [22, 23].

The fast transient EL and photo-response of the bi-functional diodes indicate their potential for application in optical communication systems. It is interesting that

the bi-functional diode in PD mode could response well to the light emitted by another identical diode in LED mode. This is due to the overlap of the light emission spectra and absorption profile as shown in Figure 2C. A visible light communication system using the bi-functional diode simultaneously as signal generator and receiver was experimentally demonstrated. The schematic set up was shown in Figure 6A. The LED mode diode was driven by a function generator while another identical diode operated in the PD mode was connected to the oscilloscope. As shown in Figure 6B–D, the light signal from the LED mode diode and the photo-response signal from the PD mode diode are compared at different frequencies. The visible communication system operated well at frequencies from 500 Hz to 2 kHz. It is noteworthy that the system operating speed is limited by the PD mode diode due to a relatively slower transient photo-response as compared to the transient EL.

This is the first demonstration of a visible light communication system using two identical CsPbBr₃ based diodes as signal generator and receiver at the same time. The bi-functional diodes with ability in broadband light detection from UV to visible range and light emission at green light are promising in multi-functional electronic applications.

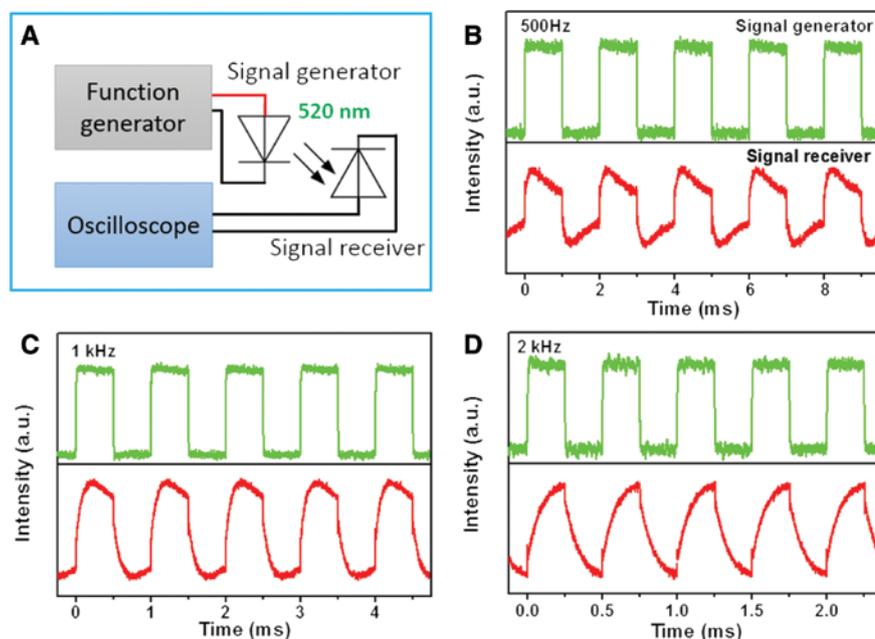


Figure 6: The demonstration of the visible light communication system.

(A) The schematic diagram illustrating the use of two identical bi-functional diodes for application in high speed visible light communication, one performed as a signal generator and another diode, placed 0.5 m away, acted as a signal receiver. The modulated EL emission and the corresponding photo-response of a pair of the bi-functional diodes, performing the visible light communication capability, at different frequencies of (B) 500 Hz, (C) 1 kHz, and (D) 2 kHz, were clearly manifested. The diode that functioned as the signal generator was operated at a forward bias of 5 V, and the one acted as the receiver was operated at 0 V.

4 Conclusions

In this work, the formation of highly reproducible and uniform CsPbBr₃ perovskite thin film layer was investigated. Our findings reveal that a uniform CsPbBr₃ perovskite thin film can be readily achieved by applying a centrifugal force during the spread of the precursor. This method avoids the ring-like inhomogeneous film morphology. The resulting highly uniform CsPbBr₃ perovskite thin films with high reproducibility were then utilized as the active layer in the diode configuration. These diodes not only emit green light with forward bias, but also exhibit UV-visible broadband light detection abilities by applying reverse bias. The bi-functionality of the diode originates from the outstanding optoelectronic features of the CsPbBr₃ perovskite active material. With fast transient EL and photo-response properties of the bi-functional diodes, a visible light communication system was demonstrated. The system employed two structurally identical bi-functional diodes, one operated in the LED mode as the signal generator and the other in the PD mode as signal receiver, respectively. This green light communication system is potentially useful in the under-water communication systems due to less absorption of sea water at this wavelength.

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