

A method towards 100% internal quantum efficiency for all-inorganic cesium halide perovskite light-emitting diodes



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ABSTRACT

An efficient all-inorganic cesium halide perovskite light-emitting diode (PeLED) was demonstrated by utilizing a mixed perovskite film of CsPbBr₃:4,5-di (9H-carbazol-9-yl) phthalonitrile (2CzPN) as the light-emitting layer, where 2CzPN was a thermally activated delayed fluorescence (TADF) material with very high internal quantum efficiency approaching to 100%. This PeLED exhibited improved EL performance with the maximum luminance of 22,063 cd/m², highest current efficiency of 8.74 cd/A and best EQE of 2.26%, which were 3.42, 4.33 and 4.35 times to that of neat CsPbBr₃ film based PeLED, respectively. The enhanced EL performance was enabled by utilization of TADF molecule as the assistant dopant that permitted efficient Förster energy transfer of all electrically generated excitons from the assistant dopant to the excited state of the perovskite emitter.

1. Introduction

Solution-processed halide perovskites have shown great potential as a new class of photoactive materials for the next-generation low-cost, high-performance optoelectronics [1–4]. Examples that have received most attention include perovskite-based solar cells with > 20% efficiency [5,6], and perovskite-based light-emitting diodes (PeLEDs) with high brightness and tunable color across the entire visible range [7–10]. Most of the premier work on PeLEDs used organic-inorganic hybrid perovskites (e.g. CH₃NH₃PbBr₃) as emitters [11,12]. Despite the demonstrated remarkable performance of the devices, the stability of organic-inorganic hybrid perovskites remains a major concern [13–15]. To avoid this problem, all-inorganic cesium halide perovskites, such as cesium lead bromide perovskite (CsPbBr₃), have been used as alternative emitters in PeLEDs [16,17]. In addition to their bright photoluminescence comparable to that of CH₃NH₃PbBr₃, cesium-based perovskites have exhibited higher thermal and chemical stability [18–20].

In terms of efficiency, for all-inorganic CsPbBr₃ perovskite LEDs, there are still many remained challenges and the potential breakthroughs. The substantial luminescence quenching found in all-inorganic perovskite CsPbBr₃ LEDs implies a fundamental conundrum for this material system in making high-efficiency perovskite LEDs [21]. Therefore, to attain high efficiencies in perovskite LEDs, the quenching

of the excitons should be minimized, kinds of methods were tried aiming to get better energy transfer in perovskite light-emitting layer. The traditional fluorescent materials of both organic polymer materials (such as poly(ethylene glycol) (PEO) [1,8], poly(N-vinylcarbazole) (PVK) [22], poly(vinylpyrrolidone) (PVP) [23]) and organic small molecular materials (such as 1,3,5-Tris(1-phenyl-1H-benzimidazol-2-yl) benzene (TPBi) [22], 1,3,5-tri(*m*-pyrid-3-yl-phenyl)benzene (TmPyPB) [24]) were involved as the assistants to transfer energy from the traditional fluorescent materials to the perovskite emitter. It is well-known that only the singlet can be harvested in the traditional fluorescent material and the triplets (three quarters of excitons) are wasted. The internal quantum efficiency (IQE) in the PeLEDs with fluorescent materials as the assistant dopant could not be larger than 25%, because of a restriction to singlet excitons imposed by spin conservation in the induced-dipole energy-transfer process, and the large ΔE_{ST} of conventional fluorescence molecules [25–28]. So, to let the triplet excitons decay radioactively in PeLEDs would be a better way to further improve the EL performance. Nowadays, newly developed thermally activated delayed fluorescence (TADF) materials are attractive for application in efficient displays. Because the metal-free TADF emitters can realize 100% excitons utilization through efficient energy transfer process of reverse intersystem crossing (RISC) from triplet excitons to singlet excitons, and are regarded as promising materials for next-generation

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OLEDs [28–32]. To date, numerous TADF emitters have been synthesized and applied in vacuum-deposited devices with commendable efficiencies. Compared with vacuum deposition, the solution process is generally accepted as a more feasible mean to realize low-cost, large-area displays or lighting products.

Thus, in this work, 4,5-di(9H-carbazol-9-yl) phthalonitrile (2CzPN), a TADF material with nearly 100% IQE, was adopted to form a composite CsPbBr₃:2CzPN film as an emitter in CsPbBr₃ based PeLEDs to harvest both singlet and triplet excitations, with significantly enhanced brightness and current efficiency. And the brightness and the current efficiency of CsPbBr₃ based PeLEDs can be remarkably improved to 22,063 cd/m² and 8.74 cd/A, respectively.

2. Experiment and measurement

2.1. Perovskite precursor solution preparation

The PbBr₂ (Alfa Aesar, 99.999%) and CsBr (Alfa Aesar, 99.999%) were mixed and dissolved in dimethyl sulfoxide (DMSO) solvent at a 1:1.2 M ratio and stirred at room temperature for 12 h in a glove box to form 10 wt% CsPbBr₃ precursor solution. 2CzPN was dissolved in DMSO solution at 0.5 mg/ml and stirred at room temperature for 12 h in a glove box. Then, 2CzPN solution and CsPbBr₃ solution were mixed with different proportions and continually stirred for 4 h to obtain a CsPbBr₃:2CzPN mixed perovskite precursor solution (The concentration of 2CzPN in the CsPbBr₃ solution were 0.01 mg/ml, 0.05 mg/ml and 0.10 mg/ml). The mixed perovskite precursor solution preparation process as shown in Fig. 1a.

2.2. PeLEDs fabrication

PeLEDs were fabricated by spin coating and thermal evaporation. Patterned indium-tin oxide (ITO, 15 Ω/square) glass substrates were cleaned successively using detergent water, ethanol, acetone, ethanol and acetone in an ultrasonic bath (each for 15 min). After 5 min treatment with ultraviolet UV-ozone plasma (120 W), PEDOT:PSS (AI4083 CLEVIOS) was spin-coated onto the ITO substrate (4500 rpm, 40 s) with a spin coater (SCO-SC100-SS) and baked at 120 °C for 20 min with a heating plate (IKA C-MAG HS7) under ambient conditions. Next, all the substrates were placed in a small transfer chamber attached to the glove box and cooled for 30 min at −1.0 bar. The mixed perovskite precursor solution was spin-coated onto PEDOT:PSS film in the glove box (H₂O and O₂ ≤ 0.1 ppm) at a speed of 4000 rpm for 60 s and immediately treated with pumping away method in the transfer chamber of the glove box with negative pressure at −1.0 bar for 20 min. The preparation of the mixed CsPbBr₃:2CzPN perovskite film was summarized in Fig. 1a.

Subsequently, the substrates were transferred into a deposition chamber (Shenyang Vacuum Technology Research Institute) which was connected to the glove box. Organic electron-transport layer (ETL) of TmPyPB and bilayer cathode Liq/Al were continuously deposited by thermal evaporation under high vacuum of $\sim 5 \times 10^{-5}$ Pa according to evaporation rates of 1 Å/s, 0.1 Å/s and 1.5 Å/s. The device active area was $\sim 2 \times 3$ mm².

2.3. Characterizations

After fabrication, we immediately encapsulated the PeLEDs with glass covers in the glove box. We used the Keithley 2400 source meter with the calibrated silicon photodetector (Beijing Normal University Photoelectric Instrument Factory, ST-86LA) to measure the current density-luminance-voltage (*J-L-V*) characteristics. The PR670 spectrophotometer was adopted to collect the EL spectra and CIE color coordinates. The photoluminescence (PL) spectra were measured by a Hitachi F-4600 fluorescence spectrophotometer. Meanwhile, the time-resolved fluorescence decay of these sample was measured with a HORIBA JOBIN YVON FluoroMax-4 spectrofluorimeter. We used the Shimadzu XRD-7000 and Shimadzu UV-2600 spectrophotometer to measure the crystallinity characterizations and Ultraviolet–visible (UV–vis) absorption, respectively. All the measurements were carried out in ambient air at room temperature.

3. Results and discussion

3.1. EL performance of PeLEDs

Using the CsPbBr₃:2CzPN composite film as the light-emitting layer, higher brightness and current efficiency PeLEDs were created. The PeLEDs adopted a typical layered architecture of ITO (120 nm)/PEDOT:PSS (30 nm)/CsPbBr₃:2CzPN X/TmPyPB (65 nm)/Liq (2.5 nm)/Al (120 nm), where “X” equals to 0, 0.01, 0.05 and 0.10 mg/ml for Devices A, B, C and D, respectively. The energy level diagram of each layer is shown in Fig. 1b, and all energy level values are taken from the literature [17,22,33–35]. The *J-V-L* characteristics are shown in Fig. 2, and the detail features are listed in Table 1. The current density increases with the doping concentration of 2CzPN increasing at each applied voltage. And all the PeLEDs with 2CzPN show improved EL performance compared to the neat CsPbBr₃ based PeLED (Device A). The Device C with 0.05 mg/ml 2CzPN in EML exhibits the best EL performance with highest luminance of 22,063 cd/m², maximum current efficiency of 8.74 cd/A, and best EQE of 2.26%, which are 3.42, 4.33 and 4.35 times to that of neat CsPbBr₃ film based PeLED, respectively. As shown in Fig. 2d, all the four PeLEDs emit bright green

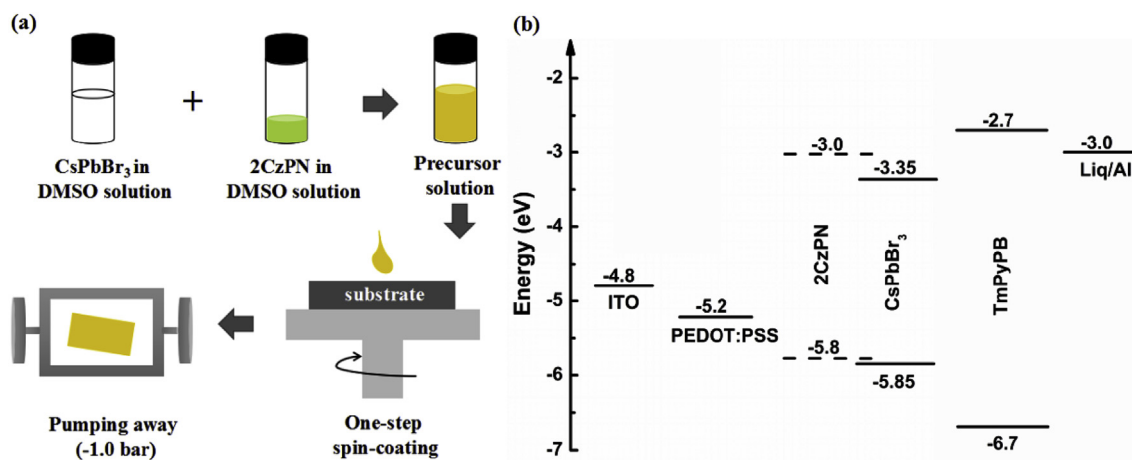


Fig. 1. (a) The schematics show the facile solution-based synthesis using spin casting followed by pumping away. (b) Energy level diagram of each layer in PeLEDs.

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