Enhanced Charge Transportation towards High Luminescent 2D Perovskite Light-Emitting Diodes

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Abstract. In this work, PEDOT: PSS 1000 was used as hole transporting layer to fabricate high luminescent perovskite light-emitting diodes. Combined with potassium doping strategy, the according perovskite light-emitting diode reaches a high brightness of 40 000 cd/m\textsuperscript{2}.

1. Introduction
Owing to their intriguing photonic and optoelectronic properties, metal halide perovskites (MHPs) are recently emerging as a promising candidate for a series of applications such as solar cells, light-emitting diodes (LEDs), photodetectors, and lasers [1-4]. In particular, due to their high photoluminescence quantum yield (PLQY), facile color tunability, solution processability, and sharp emission, perovskite light-emitting diodes (PeLEDs) have attracted significant attention [5-7]. Very recently, green-emitting, red-emitting, and near-infrared-emitting PeLEDs have obtained impressive external quantum efficiencies (EQEs) exceeding 20\%, which represents an important step toward the commercial applications [8-10].

MHPs are better known for their long carriers’ diffusion length, which is good for solar cells. However, opposite properties are desired when they are used in LEDs, i.e. PeLEDs. Thus the exciton dissociation is expected to be suppressed, in which radiative recombination will be enhanced in the perovskite films. Recently, two-dimensional (2D) perovskites have been proposed by doping large organic cations, such as PEA\textsuperscript{+} into three-dimensional (3D) perovskite frame to form Ruddlesden–Popper-type layered structure. It was found that multiple quantum wells could be assembled in these 2D perovskites, which can enlarge exciton binding energy and confine charge carriers to reduce the possibility of exciton dissociation. This makes 2D perovskites good candidate materials for efficient LEDs [8,11,12].

Nevertheless, some unfavorable characteristics have been identified, such as the undesired orientation of the layered structures, which can cause charge-transfer problems and charge accumulation [13]. In addition, solution processed thin films of 2D perovskite show reduced crystal size compared to those of 3D perovskite, which could increase the concentration of the defects and traps on the film surface and grain boundaries, leading to undesired non-radiative recombination and decreased emission efficiency [14].
In this article, potassium was introduced to 2D perovskites to passivate defects. We find the resulted films exhibit smooth, compact surface morphology with fewer observed boundaries. Bright green emission and high PLQY up to 39% were also recorded. In addition, to achieve high luminescence, a more conductive and lower work function hole transporting layer (PEDOT: PSS 1000) was introduced. Accordingly, the corresponding PeLEDs reaches a high luminescence with maximum value of 40 000 cd/m².

2. Results and Discussion

2.1. Optical properties

2D perovskites were fabricated using a one-step solution-based process. Their compositions were controlled through the ratios of PEA\textsubscript{Br}: FABr: PbBr\textsubscript{2} in dimethyl sulfoxide (DMSO) and a targeted average perovskite formula of PEA\textsubscript{2}FA\textsubscript{5}Pb\textsubscript{6}Br\textsubscript{19} was gently accomplished.

It is documented that the introduction of potassium (K) additives could efficiently passivate surfaces and stabilize luminescence across a range of bandgaps without compromising charge transport or extraction. We then synthesized K\textsuperscript{+} incorporated 2D perovskites. For K\textsuperscript{+} intercalation, KBr, with the same molar concentration as perovskite, was first dissolved in DMSO. Then, the KBr and perovskite precursor were mixed with the volume ratio of 1:9. In order to study the passivation effect of K\textsuperscript{+}, steady PL and PLQY were performed on K\textsubscript{+}PEA\textsubscript{2}FA\textsubscript{5}Pb\textsubscript{6}Br\textsubscript{19} and PEA\textsubscript{2}FA\textsubscript{5}Pb\textsubscript{6}Br\textsubscript{19}. As can be seen in Figure 1, the K\textsuperscript{+} doped 2D perovskite film show much higher PL intensity and the PLQY soars up to 39%. This demonstrates substantial mitigation of non-radiative losses after doping with K\textsuperscript{+} in our perovskite system.

![Figure 1 PL of 2D perovskites with and w/o K doping](image_url)

2.2. Morphology

The morphology of perovskite thin films is another important factor governing the luminescence property and final performance of PeLEDs. Therefore, we used a scanning electron microscope (SEM) to characterize the morphology of perovskite thin films. Figure 2a shows that the pristine PEA\textsubscript{2}FA\textsubscript{5}Pb\textsubscript{6}Br\textsubscript{19} film has a full coverage but rough morphology. We also examined the crystallinity of perovskite film with K\textsuperscript{+} doping. In contrast, as shown in Figure 2b, a dense and uniform morphology
was obtained, reflecting the doped K⁺ doesn’t disturb the crystallinity of our 2D perovskite in the film fabrication process.

![Figure 2 SEM images of 2D perovskite films (a) with and (b) w/o K doping](image)

2.3. Device performance

We further fabricated PeLEDs using K doped 2D perovskites. As indicated in Figure 3a, we configured PeLEDs as the structure: ITO/PEDOT: PSS (40 nm)/perovskite film (150 nm)/TPBi (30 nm) /LiF (1 nm)/Al (100 nm). Both devices emitted green light with the emission peak around 530 nm, which is consistent with their photoluminescence (Figure 1, 3b). Moreover, the devices exhibit narrow emission with the FWHM around 28 nm (Figure 3b). However, as shown in Figure 3c, the PeLED using PEDOT: PSS 4083 as HTL showed moderated luminescence, only recording a maximum luminescence of 7000 cd/m². This could be ascribed to the low transportation of hole carriers, which leads to mismatch with injected electrons in perovskite layer. In contrast, the device basing on PEDOT: PSS 1000 presents outstanding performance, reaches a brightness of 40 000 cd/m².

![Figure 3 Device structure and performance. (a) Device structure of 2D PeLEDs; (b) EL spectra of the PeLEDs. Inset: photograph of EL; (c) J-V-L characteristic curves of the PeLEDs.](image)
3. Conclusion

Briefly, 2D PeLEDs with high luminescence has been demonstrated via more conductive HTL of PEDOT: PSS 1000 and potassium doping. The resultant smooth and uniform perovskite film displays highly bright emission with PLQY up to 39%. The device using PEDOT: PSS 1000 as HTL achieves a high luminescence of 40 000 cd/m².

Acknowledgments

We gratefully thank the support from TCL China Star Optoelectronics Technology Co. Ltd.

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