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Perspective Review

Evaluating the viability of integrating single-unit perovskite light-emitting diodes into perovskite–perovskite tandem configurations

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ABSTRACT: As an inherent current-driven device, the luminous intensity of a single-unit perovskite light-emitting diode is directly proportional to the current density. However, this relationship can lead to a deterioration in the operational lifetime of the device at high current densities. In contrast, a tandem device structure, not only requires less current to achieve equivalent brightness compared to a single-unit device but also nearly achieves the combined efficiencies of each light-emitting unit. Herein, we present recommendations and protocols designed to facilitate the fabrication of all-tandem perovskite light-emitting diode, with the aim of benefiting both the research and industrial communities.

KEYWORDS: tandem, perovskite, light-emitting diode, atomic layer deposition, charge generation layer

1 Introduction

The inexpensive fabrication route, combined with the unique optoelectrical properties of metal halide perovskites, has made them an ideal candidate for use as an emission layer (EML) in light-emitting diode (LED) applications [1]. Thanks to the narrower full width at half maximum (FWHM) of perovskite materials, perovskite light-emitting diodes (PeLEDs) achieve improved color quality and excellent efficiency. For instance, the emission spectrum of a perovskite material is concentrated around a specific wavelength with minimal spread across different wavelengths, resulting in higher color purity. Additionally, photons can be emitted in a specific direction and wavelength range, rather than being scattered or absorbed within the perovskite material. As a result, the energy of the emitted photons is more consistent and less dispersed. These factors not only increase the proportion of photons escaping the device and contributing to overall brightness, but also reduce energy losses, ultimately enhancing the power efficiency of the devices (i.e., single junction and tandem PeLEDs). To date, the most efficient and stable EML for PeLEDs have been prepared from the perovskite precursor salts using solution-deposition method, with the crystallite size ranging from micrometres to a few nanometres [2]. However, the directly proportional relationship between the luminous intensity and current density in PeLEDs can compromise device performance and stability. Taking the inspiration from tandem organic light-emitting diode (T-OLED), combining the single-unit OLEDs to form a tandem device can overcome these issues, as it requires less current to achieve equivalent brightness compared to a single-unit device while achieving the combined efficiencies of each single-unit device [3]. Unlike well-established T-OLEDs prepared via vacuum deposition method, solution-processed PeLEDs, which are fabricated using highly polar solvents such as N,N-dimethylformamide (DMF) or dimethyl sulfoxide (DMSO), face the limitations in tandem device structures (or known as perovskite/perovskite tandem light-emitting diode (PPTLED), Fig. 1(a)). This is because the strong polar solvents can redissolve the underlying layer if no appropriate protection is applied during the deposition of the top perovskite EML. For these reasons, research publications specifically focusing on all-T-PeLEDs are yet to be widely reported.

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2 Research status of the perovskite-based tandem light-emitting diode

2.1 Perovskite/organic hybrid tandem light-emitting diode

Due to technical difficulties, researchers have focused their attention

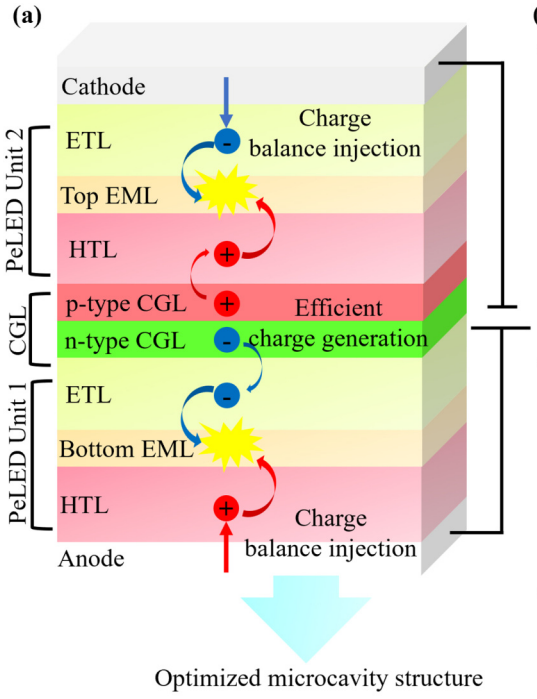

Figure 1 (a) Schematic illustration and (b) the challenges that need to be addressed to facilitate the realization of the PPTLEDs. E_1, E_2, ETL, and HTL represent LUMO of n-CGLs, HOMO of p-CGLs, electron-transporting layer, and hole-transporting layer, respectively.

on tandem devices that combine perovskite EMLs with organic EMLs, also known as perovskite/organic hybrid tandem light-emitting diode (POTLED). This structure can (i) prevent the degradation of the bottom perovskite, (ii) maintain the optoelectronic properties of the bottom perovskite, and (iii) avoid the redissolution issue of the bottom perovskite EML, as the subsequent functional layers after perovskite EML are deposited using vacuum deposition techniques. Benefitting from this, the overall performance and stability of the POTLEDs are significantly enhanced compared to that of single-unit LED (i.e., PeLED and OLED). As of now, Shanghai University in China has achieved an external quantum efficiency (EQE) of over 40% for green-POTLED (i.e., electroluminescence peak at 516 nm), along with high operational stability, with a device half-life (T_{50}) of 42,080 h at an initial luminance of 100 cd/m^2. It is important to highlight that this represents the highest EQE reported to date.

This performance was realized through the optimization of the charge generation layer (CGL), which facilitated efficient opto-electric coupling and reduced Joule heating within the device [4].

2.2 Perovskite/perovskite quantum dot hybrid tandem light-emitting diode

As noted earlier, the primary technical challenge in realizing PPTLEDs is protecting the underlying bottom perovskite EML during the subsequent deposition of the top perovskite EML using highly polar solvents such as DMSO or DMF. Benefitting from the orthogonal solvent for preparing perovskite precursor, several significant studies have offered valuable insights into the development and potential of PPTLEDs. These insights have been gained through the strategic combination of perovskite thin films (serving as the bottom EML, DMSO or DMF) with perovskite quantum dots (serving as the top EML, n-octane). However, the EQE of the resultant device is significantly lower than the reported EQE of over 40% for POTLEDs, being limited to less than 2%. This discrepancy can be attributed to (i) the inappropriate choice of materials and the method preparation for the n-type and p-type CGLs and (ii) the improper energy level alignment between the n-type and p-type CGLs, thereby restricting the charge injection efficiency into the adjacent perovskite EMLs [5, 6].

3 Realization of perovskite/perovskite tandem light-emitting diode

Since the first report of utilizing atomic layer deposition (ALD) method to protect the underlying perovskite absorber layer during the deposition of the top perovskite absorber layer using a similar solvent (i.e., a mixture solvent of DMSO:DMF), this method has been widely applied in perovskite-perovskite tandem solar cell (PPTSC) applications [7]. Leveraging the identical device structure for both all PPTSCs and PPTLEDs, where the interconnecting layer is sandwiched by the top perovskite film and the bottom perovskite film, PPTLEDs could also be realized if (i) the ALD method is used to prepare the CGL for protecting the underlying perovskite EML and (ii) the associated challenges highlighted in Fig. 1(b) are effectively addressed.

3.1 Charge generation layer

As a critical component within tandem device, the interconnecting layer—commonly referred to as the CGL—is essential for generating free charge carriers at the interface between the n-type and p-type materials. Typically, the n-type material functions as an internal cathode, while the p-type material serves as an internal anode. Upon influence of an electric field to the tandem device, free electrons can transfer from the highest occupied molecular orbital (HOMO) of the p-type material to the lowest unoccupied molecular orbital (LUMO) of the n-type material, thereby creating a hole in the p-type material. This process results in the formation of an electron-hole pair at the interface. Under the influence of the

electric field, these electron-hole pairs are dissociated into free electrons and free holes, which are then injected into the adjacent perovskite EML. To ensure efficient charge generation and facilitate effective charge injection into the perovskite EML, the energy level difference between the LUMO of the n-type CGL and the HOMO of the p-type CGL, denoted as ΔE, should be ≤ 0.5 eV [8]. Conversely, an excessively large ΔE reduces the probability of electron-hole pair formation at the interface between the n-type and p-type materials, thereby diminishing free charge generation and limiting efficient charge injection into the adjacent perovskite EML. Benefitting from the identical device structure for both PPTSCs and PPTLEDs, the CRL comprising C_{60}/ALD-SnO_x/PEDOT:PSS in PPTSCs could be adapted as a CGL in PPTLEDs. However, the large energy offset between the LUMO level of ALD-SnO_x (i.e., ~ 3.9 eV) [9], and the HOMO level of PEDOT:PSS (i.e., ~ 5.2 eV) [10] may limit the charge generation efficiency at the interface between the n-type and p-type materials, thereby deteriorating the EQE of the PPTLEDs. Therefore, optimization of these materials (i.e., interface engineering, dopant engineering, chemical modification, substituting PEDOT:PSS with alternative hole-transporting materials that possess a shallower HOMO level, etc.) is necessary to achieve a ΔE of ≤ 0.5 eV. Furthermore, C_{60} may affect the transmission of light in the visible region. Therefore, replacing C_{60} with the well-established electron-transporting materials (i.e., TPBi, Po-T_2T, 3TPYMB, TmPyPB, and B_3PyMPM) is necessary not only to reduce the electron injection barrier to the perovskite EML, but also to improve the light-outcoupling efficiency of the devices.

3.2 Bottom perovskite emission layer

The thermal stability of the bottom perovskite EML in PPTLEDs is also a critical determinant of overall device performance. Given that the EQE of the single-unit PeLEDs is highly sensitive to temperature [11], it is essential to identify perovskite EML materials capable of withstanding elevated temperatures to prevent degradation during the ALD process. Among various perovskite materials, inorganic perovskites have demonstrated superior resistance to moisture and temperature for solar cell applications [12]. Therefore, we propose that inorganic perovskites, such as $CsPbX_3$ (where X = Cl, Br, I), will become more promising candidates for use as the bottom perovskite EML compared to organic perovskite EMLs (i.e., $MAPbX_3$ and $FAPbX_3$) for PPTLED applications. We believe that selecting appropriate perovskite materials to function as the bottom perovskite EML and optimizing their physical/chemical properties will help to mitigate degradation during the subsequent ALD process for depositing ALD-SnO_x at an elevated temperature.

3.3 Microcavity effect

Since the stacked structure increases the complexity of device design and affects the optical properties of the devices, precise design considerations that account for microcavity effects and outcoupling efficiency—through optical simulation and analysis—are essential to optimize the optical properties of PPTLEDs [13, 14]. By manipulating the microcavity effect in PPTLEDs, the spectral width, emission direction, and light extraction efficiency can be optimized, thereby yielding higher efficiency and electroluminescence lifetime.

4 Conclusion and future outlooks

As of now, research on PPTLEDs, particularly those based on thin-film architectures, has garnered less attention compared to POTLEDs, probably due to the technical challenges highlighted in Fig. 1(b). A comparison between the PPTLEDs and POTLEDs in terms of their strengths, limitations and implementation barriers is presented in Table 1. Because PPTLEDs is still in an early stage of development, we advocate for the transparent reporting of device failures across key performance metrics. Such transparency is crucial for identifying and addressing the underlying issues, thereby facilitating the development of new research directions in PPTLEDs. Once PPTLEDs are optimized to achieve EQEs and device longevity comparable to those of POTLEDs, they are expected to outperform POTLEDs. This potential superiority is attributed to the significantly narrower FWHM and low-fabrication cost of PeLEDs compared to OLEDs (i.e., PPTLEDs versus POTLEDs), which enable PeLEDs to deliver sharper, higher-contrast images at a lower cost, making them a more attractive option for high-performance display and lighting applications (i.e., color-tunable emission and white light sources), with possible future applications in indoor optical communication [15]. Meanwhile, driving voltage management is also crucial for realizing high efficiency PPTLEDs. Generally, each emission layer in a PPTLED requires a specific voltage to initiate and sustain electroluminescence. In a tandem device, the driving voltage is defined as the sum of the individual voltages required by each sub-cell. If one sub-cell receives too much or too little voltage, it can lead to non-uniform emission, reduced efficiency, or even device failure. Therefore, this challenge deserves further attention in PPTLED applications as well. It is also suggested that a thermally evaporated perovskite EML can also be used to deposit on top of the solution-processed perovskite EML to form PPTLEDs for addressing the solvent orthogonality issue [16]. However, this approach might lead to uncontrolled stoichiometry of the perovskite materials and would require further optimization to ensure uniform composition and device performance.

Data availability

Not applicable.

Table 1 Comparison between the PPTLEDs and POTLEDs in terms of their strength, limitation and implementation barrier

Aspect	PPTLEDs (perovskite–perovskite)	POTLEDs (perovskite–organic)
Strengths	Narrower FWHM and higher color purity than most organic emitters	Well-established organic hole- and electron-transport materials and device architectures
Limitations	Ion migration and phase segregation under applied electric field, especially in mixed-halide blue perovskites	Organic layers are intrinsically sensitive to moisture/oxygen
Implementation barriers	Need for dense and compact CGL layers prepared via the ALD method to protect underlying perovskite, which could deteriorate the underlying perovskite EML if not handled properly	Complex multi-layer vacuum deposition lines; high-cost production

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

E. L. L.: Data collection and curation, writing manuscript, project administration, funding acquisition, supervision. X. C.: Visualization. Z. H. W.: Check manuscript, project administration, funding acquisition, supervision. All the authors have approved the final manuscript.

Use of AI statement

None.

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