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# Performance improvement based on latitude classification of perovskite light-emitting diodes

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**Abstract.** The Perovskite Light-emitting Diode (PeLED) can effectively convert light energy and electrical energy, and the study of Light-Emitting Diode (LED) is conducive to the efficient use of energy. Starting from the dimension classification of perovskite light-emitting diodes, this paper introduces the advantages of perovskite in different dimensions and the methods to improve the performance of perovskite light-emitting diodes. It is expected to realize the preparation of low-cost and high-performance perovskite light-emitting diodes. Light-emitting diodes or electroluminescent devices have many excellent properties such as high brightness, wide colour gamut, low power consumption, long life and environmental protection. They have been widely used in the field of display and lighting, and have become one of the most competitive products in the optoelectronic industry. With the development of the LED industry and the higher requirements for LED display in the new era, scientific researchers' exploration of new electroluminescent materials has also been gradually strengthened. Among them, organic molecules and new low-dimensional halogenated perovskite have attracted much attention because of their many advantages. The performance of LED depends on the type of light-emitting material and device structure. It is also important to understand the light-emitting mechanism of such devices and their internal carrier transport mechanism. Studying the light-emitting mechanism and carrier transport mechanism of LEDs based on different light-emitting materials is not only of scientific significance, but also can provide a reliable theoretical basis for further improving their performance.

**Keywords:** perovskite light-emitting diode, classification of perovskite LED, EQE.

## 1. Introduction

### *1.1. Introduction of LED*

*1.1.1. Basic structure of LED.* Although traditional inorganic LED and OLED and PeLED are electroluminescent devices, their structure is still somewhat different. The luminescent layer of the former is a pure inorganic direct bandgap semiconductor composed of materials such as N, P, As or Ga, which is essentially a pn junction diode. Under the action of forward voltage, a small number of injected carriers recombine with most carriers near the pn junction to form excitons and release excess energy, that is, emit photons. OLED and PeLED devices take organic film or perovskite film as the light-emitting layer, and their structure is similar to layered sandwiches. As shown in Figure 1, the whole device

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structure uses glass as the substrate, and organic matter or perovskite is sandwiched between the p-type empty six transmission layer and the n-type electronic transmission layer as a light-emitting layer. The two electrodes of the device are transparent anode and a metal cathode, respectively. In order to improve the performance of the device, sometimes electron layers.

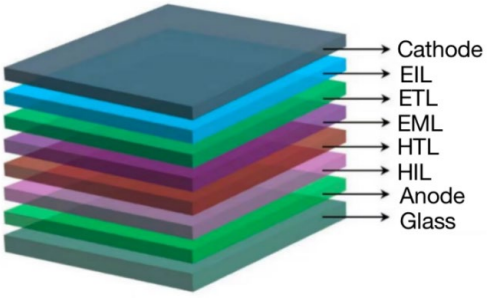


**Figure 1.** Basic structure of LED [1].

*1.1.2. The principle of LED luminescence.* The basic principle of LED luminescence is shown in Figure 2. Under the action of external voltage, the carrier is injected into the device and spontaneous radiation is generated to emit photons. After the carrier is injected through the electrode, the electrical emission in the device mainly has the following ways: 1. electrons and holes are injected into the same nanocrystal to radiate composite luminescence; 2. The injected electrons and holes form excitons in the transport layer, and then transmit energy to the transmitter through Förster energy transfer. Nanocrystals in the optical layer material produce new excitons (i.e. electron-hole pairs) and emit photons; 3 excitons in the transport layer do not transfer energy to the light-emitting layer through Förster energy transfer, but radiation in the transport layer. The photon energy emitted by it and the transmission layer material The energy level of the material is related.

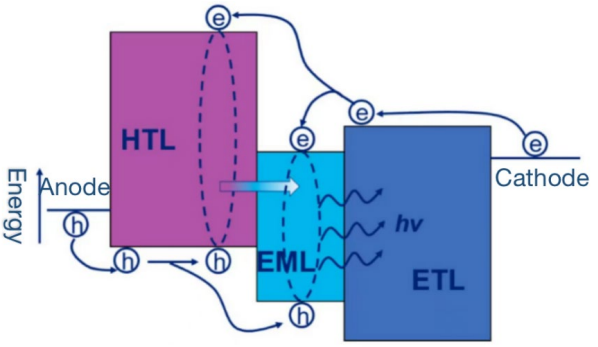


**Figure 2.** The principle of LED luminescence [1].

*1.2. Introduction of low-dimensional perovskite materials*

*1.2.1. The structure of perovskite.* The first perovskite is $GaTiO_3$, which structure is $ABX_3$. Later, researchers referred to substances with the same structure as perovskite. The structure is shown in Figure 3. A is a cation with a larger radius, B is a metal cation with a smaller radius (usually a transition metal), and X is an anion. A ion is filled between the $[BX_6]^+$ octahedral structure composed of B and X. By replacing A, B and X ions, different types of perovskite can be formed.

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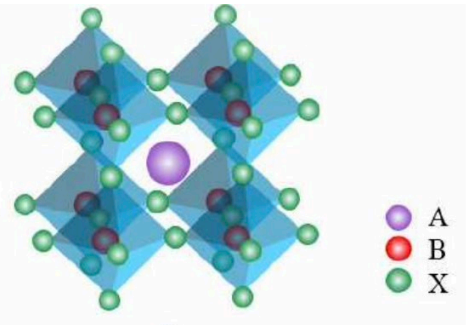


**Figure 3.** The structure of perovskite crystal [1].

*1.2.2. The classification and properties of perovskite.* Through the control of the preparation process of perovskite, different dimensions of perovskite can be produced, including zero-dimensional quantum dots, one-dimensional nanowires and two-dimensional nanochips (Figure 4).

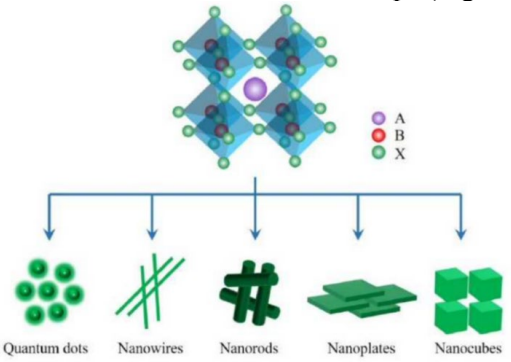


**Figure 4.** Crystal structure and morphology of low-dimensional perovskite materials [1].

Perovskite is a direct band gap semiconductor. Low-dimensional perovskite material has the advantages of high luminous intensity, adjustable size, band gap and luminous color, high absorption coefficient and high carrier mobility. It has developed into a new generation of light-emitting materials. And it has remarkable application prospects in LED devices.

*1.2.3. Working Principle of Perovskite LED.* First, electrons and holes are injected from both ends of the electrodes of the Perovskite LED device. After that, electrons and holes are injected into the perovskite thin film through the electron transport layer and the hole transport layer, respectively. In perovskite thin films, electrons and holes recombine to produce excitons, which undergo radiation transitions to produce photons, resulting in luminescence phenomena.

The lower highest occupied molecular orbital (HOMO) of the electron transport layer and the higher lowest unoccupied molecular orbital (LUMO) of the hole transport layer can act as holes and electron barrier layers, respectively, so the electrons and holes injected into the perovskite thin film can be effectively limited in the perovskite luminescent layer, which means the luminous intensity can be maximized.

*1.2.4 Three different ways of recombination.* When the exciton binding energy in perovskite thin films is relatively large, electrons and holes tend to form Frenkel excitons, which means that the main contribution of radiation recombination is single molecule recombination, with a faster recombination rate.

In thin films with low exciton binding energy, the main mode of radiation recombination is bimolecular recombination, with a slower recombination efficiency.

Auger recombination refers to when the carrier concentration in the perovskite thin film is large enough, the energy generated by the recombination of electrons and holes is transferred to another

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electron or hole, thereby stimulating it to a higher energy level. Auger recombination can reduce the efficiency of electron and hole radiation recombination in PeLED devices, so it should be avoided as much as possible when preparing PeLED devices.

According to the classification of spatial dimensions, perovskite can generally be divided into the following four categories: 0D (quantum point QD), 1D (nanometers, nanorods, nanocolumns, etc.), 2D (layered films) and 3D (body-phase films). Due to the structural limitations of 1D nanometer perovskite materials, the research mainly focuses on 2D, 3D perovskite film and 0D perovskite QD.

## 2. Different methods to improve the properties of types of perovskite LEDs

### *2.1. 3D perovskite film LED*

*2.1.1. Adjustment of light-emitting color of the device.* The use of mixed halide perovskite materials not only adjusts the light-emitting color of the device by adjusting the forbidden band width of halide perovskite materials, but also improves the efficiency of the device. This shows the advantages of the adjustable band width of mixed halide perovskite materials and the diversity of halide composition content in LED, which provides certain guidance for adjusting its luminous performance by changing the halide composition and content in the later stage.

In September 2014, 3D perovskite LED devices were prepared for the first time in an atmospheric environment. Friend et al. used a one-step solution rotary coating to prepare CH3NH3PbBr3 material as a green light-emitting layer, using the inverted structure ITO/PEDOT: PSS/$CH_3NH_3PbBr_3$/F8/Ca/Ag. At this time, the F8 polymer, as a barrier for electron transmission, effectively prevents emission quenching near the Ca metal interface. When the current density was 123 mA/cm$^2$, 0.1% green light External Quantum Efficiency (EQE) and 0.4% Internal Quantum Efficiency (IQE) were achieved. In order to adjust the luminous performance of the device, they mixed $CH_3NH_3PbBr_3$ and $CH_3NH_3PbI_3$ precursor solutions with a 2:1 molar ratio, and prepared the mixed halide perovskite luminescent material $CH_3NH_3PbBr_2I$, which glowed red in the same device structure. $CH_3NH_3I$ and $PbCl_2$ were dissolved by a molar ratio of 3:1, and the mixed halogen $CH_3NH_3PbI_{3-x}Cl_x$ perovskite precursor solution was prepared. $CH_3NH_3PbI_{3-x}Cl_x$ perovskite material was prepared by solution method as a light-emitting layer. $TiO_2$ worked as the electron transport layer, choose the F8 polymer layer with deep ionisation potential and weak electron affinity to form a heterojunction with perovskite, so that the hole was limited to perovskite, and the electrons were blocked by it and cannot flow out through the anode. When the current density was 363 mA/cm$^2$, the device with the structure ITO/$TiO_2$/$CH_3NH_3PbI_3$-xClx/F8/$MoO_3$/Ag obtained 0.76% of red light EQE and 3.4% IQE (Figure 5) [2].

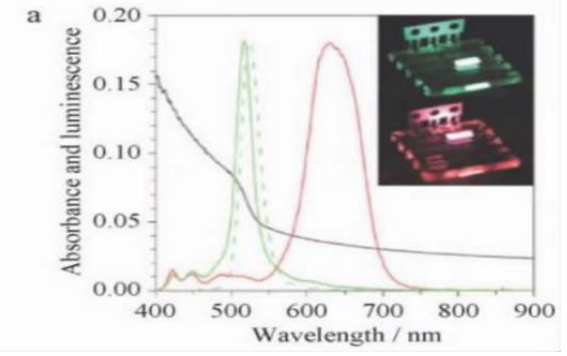


**Figure 5.** The UV absorption (black) and EL spectrum (green) based on $CH_3NH_3PbBr_3$ LED, the EL spectrum (red) based on $CH_3NH_3PbBr_2I$ LED, work schematic diagrams based on $CH_3NH_3PbBr_3$ (green) and $CH_3NH_3PbBr_2I$ (red) LED device [2].

*2.1.2. Component engineering and solvent engineering to improve the perovskite film.* In view of the problems of poor film formation and high defect density in three-dimensional perovskite, the internal

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grain size of perovskite film can be significantly changed by component engineering and solvent engineering regulation of perovskite film forming process [Figure 6 (a)], so as to obtain three with high coverage and high fluorescence quantum efficiency. Vitamin titanium ore thin film (vitaminium). The luminescent device prepared based on this film can still be maintained at a high current density of 300mA·cm$^{-2}$ external quantum efficiency. In addition, the surface of perovskite film is passivated by hydrophobic organic amine. It can block the penetration of water molecules into perovskite films and inhibit the migration of ions in perovskite films, and increase the life of the device to 23.7 h under the condition of accelerated ageing with a high current density of 100 mA·cm$^{-2}$[2].

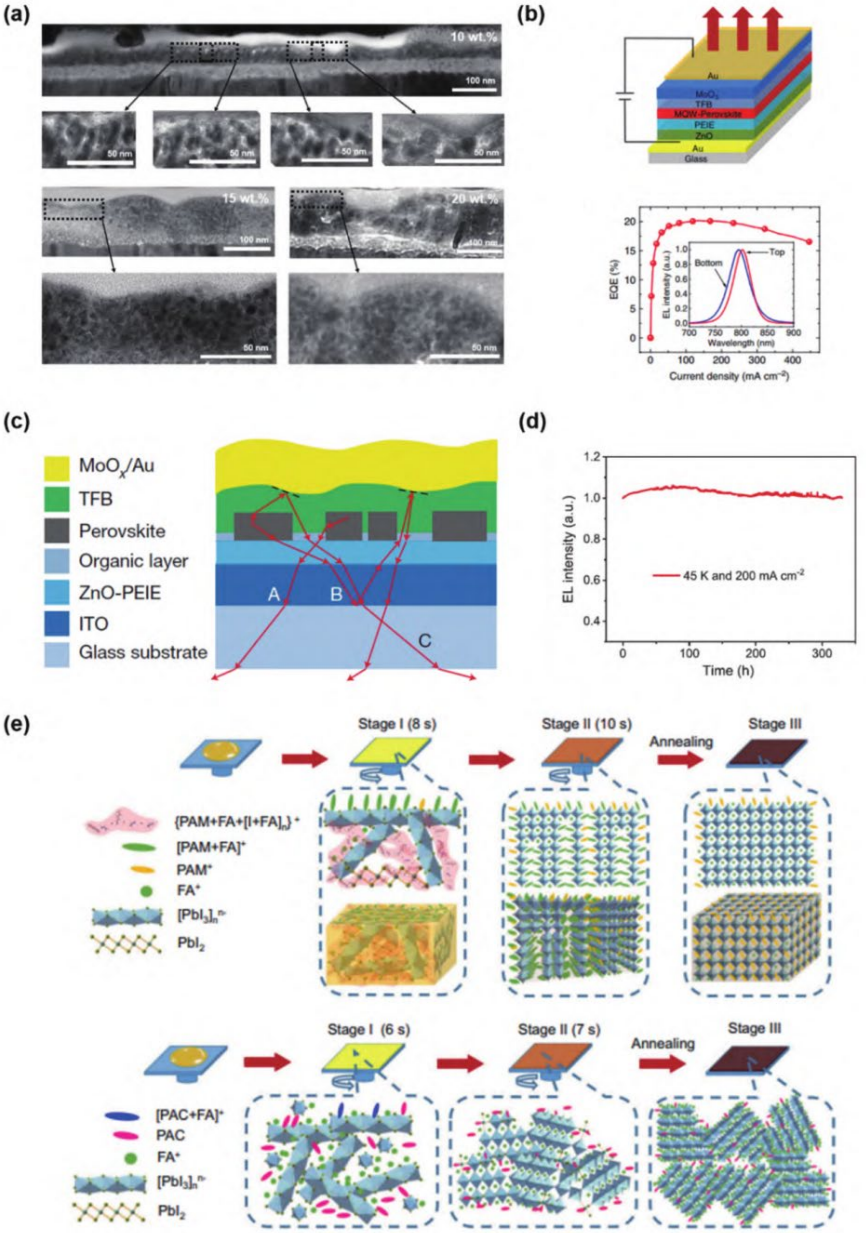


**Figure 6.** Nearinfrared threedimensional perovskites. (a) Grain distribution in perovskite films with different concentration [3]; (b) structure and performance of topemitting perovskite LED [4]; (c) schematic of device structure and light output with submicrometrestructured perovskite film [5]; (d) operation stability of devices at 45 K with a constant current density of 200 mA· m$^{-2}$ [6]; (e) schematic of growth pathways of perovskites with different additives [7].

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### *2.2. 2D perovskite film LED*

*2.2.1. The formation of quantum well structures.* Multi-layer quasi-two-dimensional perovskite compounds were synthesised by doping with large-body positive phenylethyl lammonium (PEA = $C_8H_9NH_3$), which is the first attempt of two-dimensional perovskite in luminescent devices. Because PEA has a large ion radius, it is not suitable for the halogenated lead octahedron 3D frame shared by angle, which will cause the perovskite structure to be separated into a layer form. The thickness of each cell of quasi-two-dimensional perovskite would be expanded by adding other cations. Through this formula, scale modulation can be realized by the comparison of methyl iodide methyl ammonium and PEA iodide (PEAI). The final structure is formed as PA2 $(CH_3NH_3)_{n-1}PbnI_{3n+1}$. The EQE of the near-infrared light-emitting device obtained from this structure had been upgraded to 8.8%, the radiation was 80 $W \cdot sr^{-1} \cdot m^{-2}$ (Figure 7).

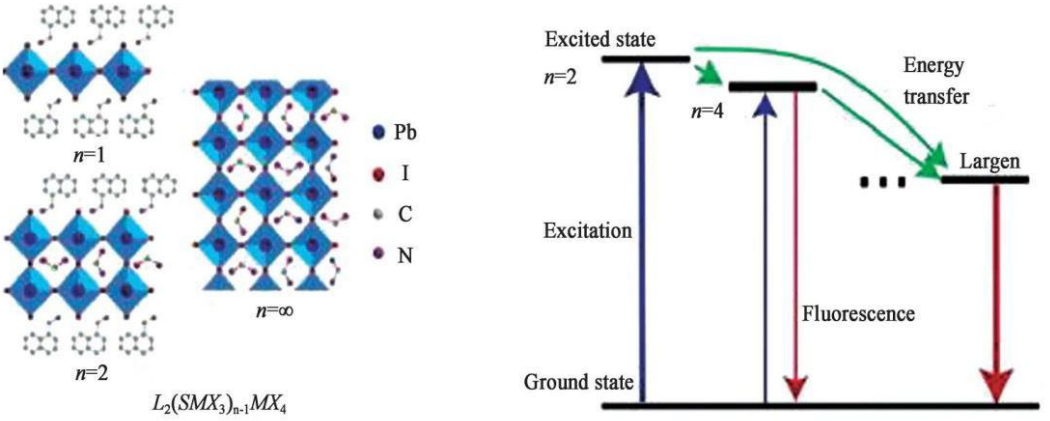


**Figure 7.** Schematic diagram of energy transfer in a two-dimensional perovskite multiple quantum well structure [2].

*2.2.2. The solvent vapor annealing method.* PLED based on pure 2D or quasi-2D perovskite materials not only has high luminous brightness and adjustable wide spectrum color, but also has good color purity. Its performance is generally better than that of 3D PLED devices. The main reason is that its "quantum well" structure significantly increases the binding energy of excitons, and the electrons and holes can be effectively limited to a small space, thus greatly improving photoluminescence quantum yield (PLQY). At the same time, the larger exciton combination can enable it to achieve efficient electron-hole composite luminescence under small light excitation or electric excitation power. By studying the proportion of organic cation components in 2D perovskite materials or controlling the growth of perovskite films by means of solvent steam treatment, the luminous performance of the device has been greatly improved.

The grain size of perovskite material is improved by solvent steam annealing method, so that the crystallinity and photophysical properties are improved, and the device EQE can also be improved, and the bright purple 2D layered PLED is opened at room temperature.

$C_6H_5CH_2CH_2CH_2NH_3^+$ (PEA) was used as an organic cation, researchers prepared $(PEA)_2PbBr_4$ perovskite film by rotary coating, and then placed it in an environment containing N, N-dimethylformamide (DMF) with a temperature of 30 ℃. Medium. Subsequently, the DMF steam spread to the perovskite film. The colour of the film changed from green to purple. It was quickly taken out and annealed on a hot plate with a temperature of 100 ℃ for 10 minutes. Compared with samples that were not annealed by solvent (Figure 8), it is found that the annealed perovskite material changed from a nanometer-scale polycrystalline film to a micron-level nanochip.

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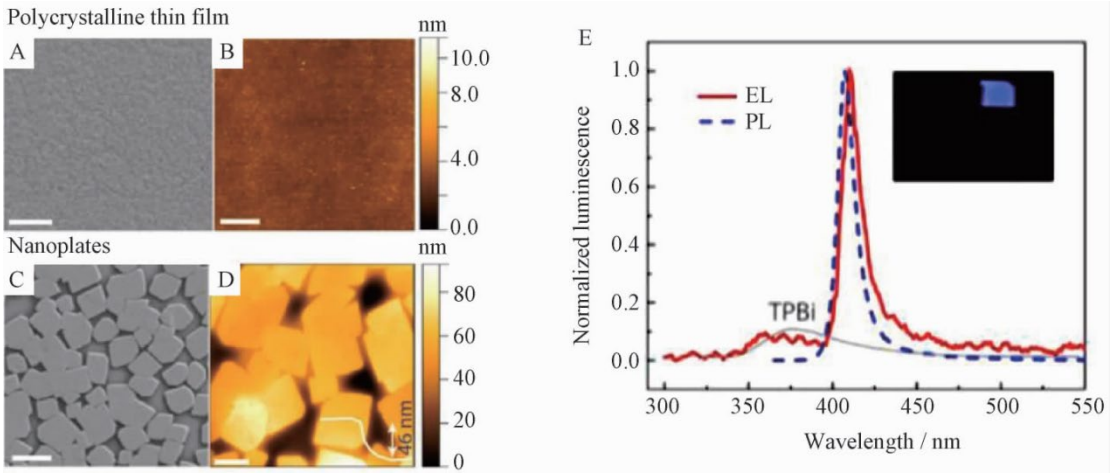


**Figure 8.** (A, B) SEM and AFM images of the $(PEA)_2PbBr_4$ thin film without DMF treatment. The scale bars are both 2 μm; (C, D) SEM and AFM images of the $(PEA)_2PbBr_4$ nanoplates obtained by annealing the thin film in DMF vapor before baking. The scale bars in panels C and D are 2and 1 μm, respectively;(E) Curves of normalized luminescence. The inset shows a picture of uniform violet light emission from a $(PEA)_2PbBr_4$ LED device [2].

### *2.3. 0D perovskite quantum dots (QD) LED*

At present, when synthesising perovskite QD, organic matter is usually used as the surface ligand of the main perovskite material, thus limiting the growth of perovskite and forming a smaller QD. Its size is limited in all spatial dimensions. It is precisely due to the influence of the quantum limiting domain effect that its PQY is relatively high. The perovskite QD used in PLED is generally an all-inorganic perovskite material ($CsPbX_3$, X = Cl, Br, I). Because of its high PLCY and tunable fluorescence emission wavelength, it shows great development potential in LEDs [8,9].

In the preparation of perovskite quantum dot devices, the synthesis of perovskite quantum dots is an important link related to the final device efficiency. At present, the two main methods for synthesizing perovskite quantum dots are thermal injection (HI) and ligand-assisted precipitation (LARP). The thermal injection method is to dissolve the oleic acid of the cation (caesium oleate Cs-OA, etc.) in the high boiling solvent octene, and inject it into the octaoctaene solution dissolved with halide ($PbCl_2$, $PbBr_2$, $PbI_2$). The precipitation method is to dissolve the perovskite precursor and long-chain organic ligand in the polar solvent dimethylformamide (DMF), and drip into a solvent with low polarity (toluene), which promotes the formation of perovskite quantum dots through the difference in solvent solubility.

An ionic exchange method was used to prepare perovskite QD, and aryl aniline hydrogen iodide (An-HI) was used to replace the commonly used long alkyl halogen ammonium salt (OAM-X, X=Cl, Br, I) or halide salt (LiX). Compared with the QD film of the original $CsPbBr_3$ and OAM-I, it is found that the anion-exchanged perovskite QD film valence band is low (OAM-I is 5.5 eV, An-HI is 5.7 eV). The ionisation energy of poly(4-butylphenylphenyl-diphenylamine) (poly-tpd) is 5.3eV. Therefore, the energy level of perovskite prepared by the anion exchange method is higher, and the hole injection barrier between poly-tpd and quantum dots is less than the original $CsPbBr_3$. The EQE peak of QD LED devices based on An-HI has reached 14.1%, CIE is (0.71,0.28). And under this condition, the EQE of QDLED devices based on OAM-I will be raised to 21.3%. Although the EQE of An-HI-based QDLED devices is lower than that of OAM-I-based perovskite QDLED devices, its operating stability is 36 times higher than that of OAM-I-based devices [10].

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## 3. Conclusion

LED technology is inseparable from human life. In recent years, with the rapid development of information technology, higher requirements have been put forward for the optical properties of the light-emitting materials used in LEDs and the preparation process of devices. This article first introduces the structure and performance of LED and perovskite, and classifies perovskite LED according to dimensions. At the same time, the improvement methods of device performance under different categories are explained. Due to the excellent performance of low-dimensional perovskite materials, LEDs based on this material are expected to develop rapidly like perovskite solar cells in the future and become a new generation of LEDs. However, there are still many factors limiting its development at present. The main obstacle is the stability and biosafety of perovskite materials. The axial integration of gradient band gap perovskite nanowires faces great challenges in a single nanostructure. This unique semiconductor nanomaterial will provide new opportunities for new integrated circuits, full-colour display, information storage and other research fields, laying an important foundation for the research and development of semiconductor integrated devices in the future.

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