Review Article

Light-Emitting Diodes Based on Two-Dimensional Nanoplatelets

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Colloidal semiconductor nanocrystals (NCs) attract significant interest in recent years due to their narrow and tunable emission wavelength in the visible range, as well as high photoluminescence quantum yield (PLQY), which are highly desired in display technologies. The high-quality NCs have been recognized as vital luminescent materials in realizing next-generation display devices. With further development, NCs with near-unity PLQY have been successfully synthesized through engineering of the core/shell heterostructure. However, as the external quantum efficiency (EQE) of the nanocrystal light-emitting diodes (LEDs) approaches the theoretical limit of about 20%, the low out-coupling factor proposes a challenge of enhancing the performance of a device when using the spherical QDs. Hence, the anisotropic NCs like nanoplatelets (NPLs) are proposed as promising solutions to improve the performance of nanocrystal LEDs. In this review, we will summarize the synthetic strategies of two-dimensional (2D) NPLs at first. Then, we will introduce fundamental concepts of LEDs, the main approaches to realize LEDs based on nanoplatelets, and the recent progress. Finally, the challenges and opportunities of LEDs based on anisotropic NCs are also presented.

1. Introduction

In the past few decades, colloidal nanocrystals (NCs) have drawn considerable attention due to their solution-processable and low-cost synthesis, high photoluminescence quantum yield (PLQY), and high color purity, as well as easily tunable emission wavelengths [1–4]. All these superior properties render NCs to offer great potential for light-emitting diodes (LEDs) [5–7]. The first colloidal quantum dot-based LEDs (QLEDs) were proposed in 1994, unlocking a new route to develop LEDs [8]. The device structure of QLEDs is similar to that of organic LEDs (OLEDs), except the emitting layer (EML) consists of QDs instead of organic materials (see Figure 1(a).) Over the past few years, the QLED technology has seen a huge development in the commercialization process. Compared with traditional semiconductor LEDs, the narrow emission makes QLEDs superior in presenting vibrant display with high saturation. A narrow emission will enable a wide color range display, which is attractive for television and other display fields [9–11]. Particularly, due to the saturated emission colors from QDs, QLEDs can cover the BT 2020 standard color parameters used for

ultra-high-definition television systems [12–13]. Therefore, QD-based displays have received a lot more attention in solid-state lighting and display technology.

With the development of device efficiency and brightness, QLEDs also face many challenges. For example, the electrical luminescence (EL) is deteriorated when the devices are operated at high current density, and the efficiency of devices is still behind the current state-of-the-art technologies. Among many properties affecting the performance of QLEDs, the external quantum efficiency (EQE) defined by the extracted photons per injected charge is the most important and can be directly compared between different devices. The EQE consists of two parts: the internal quantum efficiency (IQE) and the out-coupling efficiency. The IQE is defined by the percentage of radiative recombination events among total recombination events or, in other words, ratio of emitted photons over injected charges. Therefore, QDs with high PLQY are more likely to reach high IQE. However, high PLQY does not necessarily guarantee high IQE since the PLQY is usually measured by optical pumping in QDs which differs from the electrical pumping in QLEDs. The

FIGURE 1: (a) A schematic device architecture of typical NPL-LEDs. (b) NPLs with core/shell and core/crown structure.

latter can cause unbalanced charge injection rates between electrons and holes, resulting in charged QDs and nonradiative Auger recombination [10, 14]. Though the best IQEs from QLEDs have reached near 100%, the EQE is still limited to a maximum value of $\sim 20\%$ mainly by the out-coupling factor, calculated by the extraction efficiency of photons from the device. Several mechanisms contribute to the loss of out-coupling factor, including photon absorption, Fresnel and total reflection, and horizontally propagating light which is generated by vertical transition dipole moments (TDM) and coupled to substrate, waveguide, and surface plasmon modes [15–16].

Several approaches have been developed to enhance the out-coupling factor, for example, structured substrate surface [17], patterning of the light-emitting layer [18], high refractive index substrate [19], and other methods that can enhance out-of-plane emission or suppress total internal reflection [20–23]. Due to the randomly oriented TDMs, the out-coupling factor of QLEDs based on spherical QDs is most limited by the transverse magnetic (TM) mode generated by vertical TDMs. Therefore, anisotropic semiconductor nanocrystals (aNCs) like nanoplatelets (NPLs) are utilized as emitters in LEDs to address the current difficulties. Unlike spherical QDs, aNCs exhibit strong in-plane TDMs that can double the out-coupling factor (from 0.2 to 0.4) [16, 24–25]. aNCs also have many outstanding optical and physical properties that are different from zero-dimensional (0D) QDs. The strong quantum confinement in the vertical direction of NPLs leads to thickness-dependent optical properties, for example, narrow emission, large exciton binding energy, relatively small dielectric constant, giant oscillator strength, extremely large linear/nonlinear absorption cross sections, and ultrashort radiative fluorescence lifetime [26–30]. Additionally, because of the nanostructures with an extended volume, the Auger recombination becomes unlikely. In other word, the Auger recombination of charged excitons in NPLs is suppressed by their anisotropic geometry [31–33]. All of these advantages make aNCs promising for next-generation LEDs.

Significant progress has been made for LEDs based on aNCs during the past few years. They can be mainly divided into two categories, namely, CdSe-based and perovskite-based aNCs. To improve the performance of the LEDs based on aNCs, lots of efforts have been made to material exploration,

device physics, and structure optimization. The NPLs with different heterostructures are raised to optimize the LED performance (such as core/shell and core/crown, Figure 1(b)). In this review, we will first present the fundamental concepts of LEDs. Then, we will introduce the synthetic strategies of NPLs. Next, we will summarize two main types of NPL-LEDs, including CdSe and perovskite materials. More specifically, we will focus on the design strategy, architecture of NPL-LEDs, and methods to enhance performance. Finally, we will overview the challenges and perspectives of NPL-LEDs.

2. Fundamentals of LEDs Based on Anisotropic Nanocrystals

Compared with QLED or OLED, the development of LEDs based on aNCs is still at an early stage. However, these technologies show many similarities in device structures as well as working mechanisms. For example, the emitting layer (EML) of aNC-LED is sandwiched between the electron transport layer (ETL) and the hole transport layer (HTL), which is similar to QLED and OLED. Different from QLED or OLED, aNC-LEDs use NPLs as the emitters for their outstanding properties like narrow emission spectrum and large surface area. In terms of the ETL and HTL, their main function is to ensure that holes/electrons can efficiently transport to the EML. In order to confine the electrons within the EML and reduce the leakage electron flow, the ETL with the shallow lowest unoccupied molecular orbital (LUMO) or conduction band minimum (CBM) and HTL with deep highest occupied molecular orbital (HOMO) or valence band maximum (VBM) are desired [34–37]. Furthermore, a hole injection layer (HIL)/electron injection layer (EIL) is used to ensure that holes/electrons can be effectively injected into the HTL/ETL from the anode/cathode. In particular, ZnO nanoparticles (NPs) are widely used as ETL because of their high mobility ($\sim 1.3 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$), simple synthesis, and suitable CBM of 4.4 eV which matches the work function of the commonly used indium tin oxide (ITO, $\sim 4.7 \text{ eV}$) cathode [38].

The emission mechanism needs to be well understood in NPL-LEDs to improve their performance. The LED performance is commonly evaluated by several parameters, including EQE, power efficiency (PE), and emission color. The EQE can be expressed as

$$\text{EQE} = \eta_{\text{out}} \text{IQE} = \eta_{\text{out}}(r\gamma q), \quad (1)$$

where EQE is defined as a ratio of the number of photons emitted out of an LED to the number of injected electrons, IQE is the internal quantum efficiency, η_{out} is the out-coupling efficiency calculated by the fraction of photons escaping out of the device, r is the fraction of excitons that radiatively decay, γ represents the charge balance, and q represents the radiative recombination efficiency of the emitter. η_{out} is restricted by the total internal reflection due to the large difference in refraction indices of the different layers in LEDs. For most QLED, the η_{out} is normally limited to 0.2 which further restricts the EQE to ~20% [3, 14, 39–41]. Several strategies have been developed to solve this issue, such as microlens array, surface plasmon resonance, and surface coarsening. However, due to the random TDM orientation in spherical QDs, QLEDs show a fixed value of the out-coupling efficiency at 0.2. Compared with QLEDs, NPL-LEDs show doubled out-coupling efficiency because of the anisotropic geometry of NPLs [42].

In addition to the light out-coupling efficiency, the IQE also needs to be increased to achieve highly efficient NPL-LEDs. A straight way to increase the IQE is to generate more photons in the NPLs by suppressing nonradiative exciton recombination. Hence, NPLs with core/shell, core/crown, or alloyed core structures are proposed. Besides, heterostructure NPLs with high PLQY and low surface defect densities are beneficial to fabricate high-quality LEDs. Controlling the shell thickness can also reduce the Auger process [43].

Furthermore, because unfavorable energy barrier exists between adjacent layers in LEDs, enhancing the charge balance is also critical to improve the performance of LEDs. Therefore, charge transport materials are the key to charge balance. There are four categories of charge transport materials used in QD-LEDs, including polymer, organic small molecule, inorganic and hybrid organic-inorganic charge transport materials [1, 44–46]. The recent emergence of organic-inorganic hybrid charge transport materials has led to the rapid development of QLEDs. For NPL-LEDs, the selections of charge transport materials are still limited. Hence, many efforts are urgently needed to improve the charge balance in NPL-LEDs. Several strategies have been reported, such as doping small amount of PVK (5 wt%) into EML [47] and introducing a PMMA layer between EML and HTL [3].

The PE can be calculated in terms of EQE and the voltage U applied to the LED: [48]

$$\text{PE} = \frac{\hbar\omega}{qU} \text{EQE}, \quad (2)$$

where $\hbar\omega$ is the average energy of emitted photons and q is the elementary charge. Obviously, high EQE and low U result in high PE where low U can be acquired through applying materials with high charge mobility or reducing the energy barriers between adjacent layers [49–50]. Finally,

the emission color determines the color gamut of displays made of NPL-LEDs which can be measured by the CIE (Commission Internationale de l'Eclairage) color coordinates. Specifically, narrower emission spectrum leads to higher color space [51]. Different from colloidal QDs which need to strictly control three dimensions for narrow spectrum, NPLs only need to precisely control the thickness. The heterostructures of NPLs with uniform thickness exhibit narrow full width at half maximum (FWHM) (~20 nm) which is highly beneficial to the color purity.

3. The Synthetic Strategies of 2D Nanoplatelets

Since the early 2000s, the anisotropic growth of NCs was first achieved for nanorods and tetrapods [52–53]. Later, it was found that the right choice of ligands could control the shape of NCs from spherical seeds to 2D NPLs [52–56]. Since the CdSe NPLs were first reported in 2008 [54], the 2D NPLs quickly received intense interest, and different strategies were developed to systematically tune the thickness, shape, and doping of NPLs [26, 57]. Heterostructured NPLs including core/shell and core/crown were also developed [30, 58–59]. During the past few years, perovskite NPLs have attracted a lot of attention. The organic-inorganic perovskite MAPbBr₃ (methylammonium (MA)) NPLs were first synthesized by Tyagi et al. in 2015 [60]. The inorganic perovskite CsPbBr₃ NPLs with high PLQY (up to 84%) were first reported by Bekenstein et al. [61] By cation exchange, CsPbI₃ and CsPbCl₃ nanoplatelets were also prepared. In recent years, the synthesis of NPLs with thickness tunability, shape control, heterostructures, and controlled doping has made great progress.

3.1. Thickness Tunability. The optical properties of NPLs are in close relation to the thickness which is defined as the number of monolayers (MLs). Typically, the optical gain threshold and Auger recombination rate are strongly correlated with the thickness of NPLs [4, 33, 62]. It has been reported that CdSe NPLs emitting at 390–550 nm, corresponding to thickness of 2–5 ML, can be synthesized by modifying the growth temperature and corresponding seed diameter [6, 54–55, 63]. Ithurria et al. studied the formation process of CdSe NPLs through a combination of transmission electron microscopy (TEM) and absorption/emission spectroscopy [55]. It was found that the growth of NPLs started from ~2 nm diameter seeds and continued by the lateral extension to form NPLs. The thickness of NPLs was determined by the seed dimension and remained unchanged during the growth. By slightly modifying the growth condition to change the size of seeds, CdSe NPLs can be grown with controlled thickness (3, 4, and 5 ML) and lateral dimension of tens of nanometers. The growth mechanism of NPLs was further studied by Riedinger et al. [64] By combining experimental and theoretical studies, it was confirmed that the growth of NPLs was mainly driven by the much lower energy required to grow on the side facets than that to create a top or bottom layer, resulting in a very large difference in growth rate between lateral and vertical directions. However, due to the difficulty for larger seeds to grow into anisotropic

FIGURE 2: (a) Reaction schematic, with a photograph of the different NPL samples under UV illumination. (b) Absorption and PL spectra of the NPL series. (c, d) BF-TEM images of 4.5 ML and 7.5 ML CdSe NPLs. Adapted with permission from reference [65]. Copyright 2018 American Chemical Society.

NPLs, little work has been done for direct synthesis of thick CdSe NPLs. Christodoulou et al. used a two-step reaction that switched from 2D to 3D growth and obtained thick (8.5 ML) CdSe NPLs with emission peak at 625 nm (Figure 2(b)) [65]. In this work, 4.5 ML CdSe NPLs were first synthesized; then, the temperature was increased to 280–320°C and chloride (CdCl₂) was introduced to stimulate the growth along the (001) direction on the top and bottom surface of CdSe NPLs to obtain the final thick NPLs (Figure 2(a)). The TEM images are shown in Figures 2(c) and 2(d). As reported by Cho et al., Cl[−] anions on the NPL can not only modify the surface energy but also reduce the nucleation barrier of CdSe islands on the basal facets [66]. Hence, the vertical growth rate was increased, resulting in more isotropic growth. This work provided a new pathway to modulate the thickness of NPLs.

For inorganic perovskite NPLs, it has been shown that CsPbBr₃ NPLs were prepared with thickness in the range of 1 to 5 MLs by varying reaction temperature (90–130°C) [62]. The same materials have also been reported by Akkerman et al. [62, 65] CsPbBr₃ NPLs can form in seconds by injecting acetone into the mixture of Cs oleate, PbBr₂, HBr oleylamine, and oleic acid. The thickness of NPLs can be controlled by the amount of HBr in the mixture. Furthermore, Zhao et al. reported CsPbBr₃ NPLs synthesized by an amino acid-mediated pathway with thickness from 4 to 12 ML by varying tryptophan (Trp) concentration [67]. The growth mechanism was proposed as the nitrogen in the indole ring of Trp attached on Pb²⁺ restraining the growth in the vertical direction, leading to the formation of NPLs. For organic-inorganic perovskite NPLs, thickness tunability and consequent changes in optical properties have also been studied. Weidman et al. synthesized $n = 1$ and $n = 2$ NPLs in the form of L₂[ABX₃] _{$n-1$} BX₄, where n represented the layers of metal halide

octahedral and was controlled by the proportion of precursor salts (AX, BX₂, and LX) added [68]. It was observed that $n = 1$ NPL had reduced PLQY compared to $n = 2$ NPLs, which was in agreement with the thickness-dependent trend.

3.2. Shape Control. Similar to the thickness tunability, the shape and area control of NPLs are also important methods to tune the properties of NPLs. The control of the NPL aspect ratio (AR) can be achieved by modifying the proportion of hydrated Cd (OAc)₂ in a Cd (OAc)₂/Cd (OAc)₂·2H₂O mixture [63]. It was shown that the AR of CdSe NPLs depends on the formation of hydroxide anions in the cadmium-acetate precursor. The CdSe NPLs can be tuned from square (AR 1 : 1) with a dry Cd (OAc)₂ precursor to elongated rectangles (AR 7.7 : 1) with a mixture of 30 mol% dry Cd (OAc)₂ and 70 mol% Cd (OAc)₂·2H₂O precursor. It was speculated that the surface energy balance can be adsorbed and altered via hydroxide anion, leading to different shapes through varying the growth rate of specific facets. This work demonstrated that the OH[−] is crucial to controlling the shape of NPLs in the growth.

Halides have been traditionally used in the synthesis of NCs to modify the shape, dimension, phase, etc. [69–70] Gerdes et al. added different amounts of halogenated alkanes (1-bromoheptane (Hep), 0.0625 to 4 mmol) to the hot-injection synthesis to tune the CdSe NPLs' shape from sex-angular to quadrangular and triangular [71]. The in situ-formed halide ions in 1-bromoheptane worked as cadmium complexing agents and surface X-type ligands to alter the nucleation and growth to change the shape of NPLs. This work also proved that different amounts of Cl-Hep and I-Hep also control the shape of NPLs. Moreover, high concentrations of Br-Hep can change the phase of NPLs from zinc blende (ZB) to wurtzite (WZ).

Apart from the well-developed metal chalcogenide NCs, shape control of perovskite NCs has also been increasingly studied in recent years. Liang et al. first reported a shape-controlled synthesis of all-inorganic CsPbBr_3 perovskite NCs [72]. Shapes of the prepared CsPbBr_3 NCs could be systematically engineered into single and lamellar-structured 0D quantum dots, as well as face-to-face stacking 2D NPLs and flat-lying 2D nanosheets via tuning the amounts of oleic acid (OA) and oleylamine (OM). The CsPbBr_3 NPLs obtained by adding 0.5 mL of OA and 0.5 mL of OM showed the PL emission peak of 449 nm. This work was significant for the application of inorganic perovskite in blue LEDs.

3.3. Heterostructures. Due to the outstanding optical features, such as narrow FWHM of PL spectrum and fast PL decay, NPLs have become promising candidates for next-generation quantum emitters [73]. For this purpose, increasing further their PLQY is critical. For QDs, the widely accepted strategy is in situ growth of heterostructures where the exciton wave function can be confined within the core by type I or quasi-type II band alignment between the core and the shell [74–75]. This can further reduce the effect of surface defects. Consequently, the PLQY and PL stability can be significantly increased. In this section, we will discuss the heterostructures of 2D NPLs including core/shell and core/crown heterostructures.

3.3.1. Core/Shell Structure. Currently, cadmium chalcogenide NPLs are still arousing the interest of many researchers due to their excellent optical performance. Table 1 summarizes the properties of several CdSe-based core/shell NPLs. Early studies often use low-temperature routes to grow NPL heterostructures due to the instability of NPLs under high temperature. Mahler et al. first reported mixing CdSe NPLs with thioacetamide (TAA), cadmium nitrate, octylamine, and chloroform to prepare the core/shell structure [30]. During the reaction, TAA and octylamine formed an ammonium sulfide complex in situ which acted as the sulfur precursor. The growth was carried out for several hours at room temperature to grow 3 to 5 ML CdZnS shells. Polovitsyn et al. reported a single-source precursor method based on zinc diethyldithiocarbamate (Zn (DDTC)_2) [76]. CdSe NPLs in hexane were mixed in toluene, followed by adding a mixture of ZnCl_2 in oleylamine, N-trioctylphosphine (TOP), Zn (DDTC)_2 , and carbon disulfide (CS_2). The reaction finished after 1 h at 110°C to grow the ZnS shell. By varying the amounts of injected precursors, a 12 ML ZnS shell can be obtained. Using 1,2-dichlorobenzene as solvent, precursors were injected by a syringe pump at 150°C and a thicker ZnS shell can be obtained. CdSe/CdS and CdSe/CdZnS nanoplatelets can be prepared by the same procedure.

Another low-temperature method for shell growth is the colloidal atomic layer deposition (c-ALD) [58]. In this procedure, the reaction is conducted in polar medium, and the sulfide source often uses the S^{2-} precursor. The growth depends on the phase transfer of the NPLs from a nonpolar phase to a polar phase and the ligand change from oleate to S^{2-} or SH^- . The ligands can then continuously react with the NPL surface through adding Cd and S precursors. This

TABLE 1: Summary of the typical properties of core/shell NPLs.

Chemical composition	Emission peak (nm)	FWHM (nm)	PLQY (%)	Ref
CdSe/CdS	586	~20		30
CdSe/ $\text{Cd}_{0.7}\text{Zn}_{0.3}\text{S}$	644		60	30
CdSe/ZnS	593-611	20-23	58-64	76
CdSe/ CdZnS	633	~20	80	78
CdSe/CdS	667	20	60-70	79
CdSe/ZnS	616	27	98	80
CdSe/CdS/ZnS	662	23.6	78	81
4-ML-CdSe/ $\text{Cd}_x\text{Zn}_{1-x}\text{S}$	655	19.5	88	81
6-ML-CdSe/ $\text{Cd}_x\text{Zn}_{1-x}\text{S}$	692	22.1	92	81
CdSe/ZnSe	684	22.1	47	81

method can also tune the properties of charge carriers which are critical for narrow band gap thin-film transistors [77].

Though the low-temperature growth of the shell can enhance the optical performance of NPLs, the PLQY is still relatively low and the FWHM is often broadened due to the nonuniform and defective growth at low temperature [30, 76, 78]. Thus, high-temperature shell growth was proposed to overcome these limitations. Rossinelli et al. reported the growth of uniform CdS shells over CdSe nanoplatelets at 300°C [79]. CdSe NPLs, Cd(oleate)_2 , and 1-octadecene (ODE) were first mixed in a round-bottom flask and heated to 80°C when oleylamine (OAm) was added. The temperature was raised to 300°C under N_2 . Next, 1-octanethiol in ODE as the S precursor was injected at a rate of 3 ml/h to prevent the formation of shells with irregular shapes. Finally, CdSe/CdS core/shell NPLs with emission at 667 nm and a narrow FWHM of 20 nm were obtained. It was noted that although OAm can etch CdSe NPLs, it can also increase the stability and PLQY of NPLs by passivation and stabilization of surface facets. The increased PL line-width (20 nm) of core/shell NPL ensemble compared with that of core CdSe NPLs was attributed to the phonon coupling caused by the strain at the interface or the nonuniform shell thickness. The optimal reaction temperature was also identified at 300°C. This method overcame the disadvantages of low-temperature growth to get high-quality products with higher PLQY and stability. Similar methods were also applied to grow CdSe/ZnS [80], CdSe/ZnSe, CdSe/ZnCdS, and CdSe/CdS/ZnCdS core/shell NPLs [81]. Altintas et al. used a modified recipe from the previous report [79] for hot-injection growth of the ZnS shell for CdSe/ZnS core/shell NPLs [80]. Zn-acetate and octanethiol were selected as Zn and S precursors, respectively. A high temperature of 573 K was required for the shell growth due to the low reactivity of octanethiol. Normalized PL and absorption spectra for CdSe/ZnS NPLs with different shell growth time (Figure 3(a)) revealed redshifted excitonic features as the thickness of the ZnS shell increases. With optimized ligand concentrations (OA and ODE), the thick-shell CdSe/ZnS NPLs exhibited near-unity PLQY of 98% (Figures 3(b) and 3(c)). Therefore, for shell growth, high temperature is considered a better strategy than low temperature for bright NPLs.

(b)

(c)

FIGURE 3: (a) Absorption and emission spectra of CdSe/ZnS core/shell NPLs during the reaction showed redshift as the shell was growing thicker. (b, c) TEM image (b) of thick-shell CdSe ZnS NPLs with 98% PLQY (c). Adapted with permission from reference [80]. Copyright 2019 Wiley-VCH GmbH.

3.3.2. Core/Crown Structure. Different from the core/shell structure where the growth of the shell occurs over all of the facets of the NPLs, the growth of the crown in the core-crown structure occurs only in the plane direction. In a typical reaction, the crown growth precursor was mixed with the Cd precursor ($\text{Cd}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$ and oleic acid) and S precursor (S-ODE or sulfur in octadecene). The mixture was then injected into the CdSe NPLs dispersed in ODE. The lateral extension of the crown can be modified by the amount of the introduced precursor. CdSe/CdS core/crown NPLs have shown high PLQY while the narrow FWHM (less than 15 nm) remained [28].

The core/crown structure has been extended to type II band alignment with a crown made of CdTe [59]. The

TOP-Te was used as the Te precursor. The electrons and holes are spatially separated between the core and the crown, and redshifted PL emission can be observed. Recently, a ternary heterostructure of CdSe/CdS/CdTe core/barrier/crown nanoplatelets was reported [82]. Here, CdSe NPL is a core, CdS is a type I barrier, and CdTe is a crown. The CdS precursor was first prepared by mixing ODE-S, $\text{Cd}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$, ODE, and oleic acid. CdSe NPLs, ODE, and $\text{Cd}(\text{propionate})_2$ were mixed and heated to 235°C when CdS growth solution and Te growth solution were added dropwise (3 ml/h) and react separately for a certain time. The CdSe/CdS/CdTe heterostructure allowed adjusting both direct and indirect transitions and even photon upconversions by tuning the barrier and crown thickness.

3.4. Doping. At present, the thickness of 2D NPLs has been perfectly controlled, and as a result, the inhomogeneous broadening of the optical features does not emerge. However, the continuous tunability of the PL emission energy is lost due to the stepwise growth. It has been demonstrated that doping is a feasible method to tune the emission color from green to red for NPLs. Sharma et al. demonstrated that introducing copper ions into CdSe NPLs led to a large Stokes shift of the PL energy from 640 to 830 nm [83].

Cation exchange has been proposed as a favorable choice for doping 2D NPLs [84]. In cation exchange reactions, host cations in the crystal lattice are replaced by minority dopant cations, resulting in interesting optical properties. Dufour et al. used cation exchange to prepare Ag^+ -doped CdSe NPLs (see Figures 4(a)–4(c)) [85]. Ag^+ cations were induced by adding silver oleate dissolved in hexane to the solution of CdSe NPLs at a rate of 6 ml/h. The TEM images of doped NPLs are shown in Figures 4(d)–4(f). The absorption spectrum remained unaffected after Ag^+ doping, while the PL spectrum changed significantly. Without Ag^+ cations, CdSe NPLs showed a narrow emission at 510 nm with FWHM of 8 nm. However, after Ag^+ doping, a broad (FWHM = 18 nm) as well as redshifted emission at 660 nm appeared (see Figures 4(g) and 4(h)). The emission color of NPLs can be tuned from green to yellow, orange, or red by tuning the amount of Ag^+ cations (see Figure 4(j)). With the continuous tunability of the emission color, this material can be applied for display applications.

Khan et al. used the same method to obtain colloidal Ag-doped CdSe NPLs [86]. The emission from 609 to 880 nm can be achieved by doping with Ag^+ . They demonstrated that lifetimes of CdSe:Ag NPLs were about twice longer than those of CdSe:Ag QDs (200 ns), and the electron-hole overlap was less than that of QDs. In addition, these CdSe:Ag NPLs had a strongly suppressed emission reabsorption due to the large Stokes shift. Different from the previous reports that reacted at or below room temperature, Galle et al. reported a seeded-growth method by injecting solutions of cadmium acetate dihydrate, selenium dioxide, and mercury acetate [87]. This reaction took place at high temperature (200°C or 240°C), and the temperature played an important role in controlling the incorporation of the Hg^+ ions; i.e., the emitters showed two red PL emission peaks at 200°C, while only one can be observed at 240°C.

Doping of NPL heterostructures have also been observed. Delikanli et al. reported the synthesis of Mn^{2+} -doped CdSe/CdS core/shell and core/multishell NPL heterostructures [88]. They grew a $\text{Cd}_{0.985}\text{Mn}_{0.015}\text{S}$ monolayer shell on undoped CdSe NPLs using the c-ALD technique. The locations of the Mn ions were controlled with atomic layer precision, which contributed to a narrow photoluminescence bandwidth of ~22 nm.

2D perovskite NPLs also exhibit interesting properties after doping. Mir et al. prepared 2.2 nm thick Mn-doped CsPbCl_3 NPLs [89]. A solution of Cs-oleate, ODE, OA, and OAM was vigorously stirred at first. Next, a mixture of PbCl_2 solution (PbCl_2 , DMSO, DMF, HCl, and HNO_3) and $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$ was injected to grow Mn-doped CsPbCl_3 NPLs. The excitonic energy can efficiently transfer to the

Mn^{2+} dopants emitting orange PL from the d electrons. In addition, due to the spin-forbidden ${}^4\text{T}^1_1\text{A}^1$ transition of the Mn d electron, the PL showed a long lifetime of 1.6 ms. Mn-doped CsPbCl_3 NPLs were further converted into Mn-doped CsPbBr_3 NPLs through anion exchange with a weaker Mn emission. Subsequently, Li et al. proposed a facile solvothermal method which can efficiently dope CsPbCl_3 NPLs with Mn. Mn could even completely replace Pb to form a new CsMnCl_3 phase by spinodal decomposition, while the host perovskite NPLs had no morphology or size change [90]. This work provided a new path to study interactions between dopant and NCs. Very recently, Davis et al. demonstrated an extremely facile ligand-mediated synthesis of 2D CsPbX_3 NPLs under ambient conditions by controlling the surface ligand composition and concentration of a mixture of short (8-carbon chain) and long (18-carbon chain) ligands [91]. This method was also applied to the synthesis of Mn: CsPbX_3 NPLs. Furthermore, the effect of Mn^{2+} ion doping on the charge carrier dynamics of CsPbBr_3 NPLs has been explored by Babu et al. [92] They found that Mn-doped CsPbBr_3 NPLs exhibited utterly fast, concurrent charge (electron) and energy transfer from the host to the dopant.

4. LEDs Based on Nanoplatelets

NPL-LEDs can be generally divided into three categories by the EML materials, namely, CdSe, all-inorganic perovskite, and hybrid perovskite NPL-LEDs. By the device architecture, these LEDs can also be grouped into two categories, i.e., conventional and inverted structures. In conventional structure, the HIL and HTL are at the bottom of the device and in direct contact to ITO while in inverted structure, the EIL and ETL are at the bottom. Both conventional and inverted structures can be applied for NPL-LEDs, and the choice of device structures is determined by the combination of material compatibility, device fabrication process, and electron/hole injection efficiency.

4.1. CdSe-Based NPL-LEDs

4.1.1. Core-Only CdSe NPL-LEDs. In 2015, Vitukhnovsky et al. reported core-only CdSe NPL-LEDs [93]. They used CdSe NPLs as emitters and organic 3-(biphenyl-4-yl)-5-(4-tert-butylphenyl)-4-phenyl-4H-1,2,4-triazole (TAZ) and $\text{N,N}'$ -bis (3-methylphenyl)- $\text{N,N}'$ -bis(phenyl)-benzidine (TPD) materials as the electron and hole transporting layers for NPL-LEDs. Since core-only NPLs have narrower PL emission than core/shell NPLs, the former can potentially offer NPL-LEDs with better color purity. In this work, the NPL-LED can exhibit an EL emission peak at 515 nm under a turn-on voltage of 5.5 V. However, compared with the PL spectrum of CdSe NPLs, the EL spectrum was significantly broadened, possibly because of the change in position of luminescence maxima of CdSe NPLs located in the vicinity of electron and hole transporting layers.

Fan et al. also demonstrated NPL-LEDs based on alloyed $\text{CdSe}_{1-x}\text{S}_x$ NPLs [7]. These alloyed NPLs showed uniform lateral dimensions and thickness of 5 to 6 ML, and the PL

FIGURE 4: Scheme of CdSe 2D NPLs (a) without and (b, c) with various amounts of Ag. (d, e, f) TEM of CdSe NPLs with an increasing amount of silver per NPL (0, 18, and 139). (g) UV spectra of CdSe NPLs for various amounts of Ag^+ doping. (h) Corresponding PL spectra of CdSe NPLs for various amounts of Ag^+ doped from 0 to ~139 silver atoms per NPL. (i) Chromaticity diagram highlighting the position associated with the PL spectra of CdSe NPLs with various amounts of Ag^+ doped. (j) Image of CdSe NPL solution with various amounts of Ag^+ doped. Adapted with permission from reference [85]. Copyright 2019 American Chemical Society.

FIGURE 5: (a) Right: schematic device architecture of the NPL-LEDs. Left: cross-sectional scanning electron microscopy (SEM) of the hybrid NPL-LED device. (b) The molecular structure for hole transport materials. (c) Energy band diagram of this device's architecture. The black arrow indicates a shift of the band edge because of the quantum confinement. (d) EQE plotted as a function of current density of NPL-LEDs based on different structures. Adapted with permission from reference [4]. Copyright 2013 Wiley-VCH GmbH.

wavelength changed (513 nm to 481 nm) with different x values (0 to 0.67). Notably, the PLQY decreased from 37% to 10% as more sulfur was alloyed, which was due to the less passivated dangling bonds from the CdS component than those from the CdSe component. Inverted device structure was used with solution-processed ZnO as ETL and 4,4'-bis(N-carbazolyl)-1,1'-biphenyl (CBP) as HTL. Similar to PL, the EL emission spectrum can also be continuously tuned by varying the Se to S ratio; for example, 496 nm, 507 nm, and 520 nm correspond to $x = 0.6$, 0.34, and 0, respectively.

These early demonstrations of NPL-LEDs based on core-only CdSe NPLs demonstrated the great potential of using NPLs for high efficiency LEDs with narrow emission bandwidth. However, compared with core/shell NPLs, core-only NPLs are less stable and often have lower PLQY. Therefore, the study of NPL-LEDs has been mostly focused on heterostructured core/shell and core/crown NPLs.

4.1.2. Core/Shell CdSe NPL-LEDs. In 2014, Chen et al. reported the first red (650 nm) LED based on quasi-2D colloidal core/shell CdSe/CdZnS NPLs [4]. In this work, the

CdZnS shell can increase the stability of luminescence properties of CdSe NPLs. The HTL was poly(vinylcarbazole) (PVK, ~20–30 nm) or poly(N,N'-bis(4-butylphenyl)-N,N'-bis(phenyl)benzidine (PTPD, ~30–40 nm) (Figure 5(b)), and the device structure was ITO/PEDOT:PSS/HTL/NPLs (30–40 nm)/ZnO (60 nm)/Al (Figure 5(a)). The energy band diagram of the LEDs is shown in Figure 5(c). Two strategies were used to enhance the device performance. To improve the charge injection and transport, the long (i.e., oleic acid) ligands of the as-synthesized NPLs were exchanged to shorter ones (i.e., 3-mercaptopropionic acid), leading to about 2-fold increase in EQE of 0.63% and a maximum luminance of 4499 cd m⁻², although the PLQY decreased by 40% after the ligand exchange (Figure 5(d)). Second, the turn-on voltage was reduced from 4.7 V to 2 V by increasing the hole mobility of HTL from ~10⁻⁷–10⁻⁶ cm² V⁻¹ s⁻¹ to ~10⁻⁵ cm² V⁻¹ s⁻¹ through replacing PVK with PTPD (the chemical structures of HTLs are shown in Figure 5(c)). However, this led to a decrease in the maximum EQE (0.28%) and luminance (2173 cd m⁻²). A possible reason was that the charge balance in PTPD-based NPL-LED was

poorer than that in PVK-based NPL-LED. Overall, the EL of NPL-based LEDs exhibited very narrow FWHM of 25 to 30 nm, indicating that core/shell NPLs have great potential in achieving pure color for LEDs.

Improved red NPL-LEDs were reported by Giovanella et al. in 2018 [42]. The LEDs used CdSe/CdZnS core/shell NPLs with 4-ML CdSe core and 3 nm CdZnS shell on each side. These NPLs exhibited PL at 658 nm and PLQY of 40%. To increase the EQE, a charge-regulating layer of conjugated polyelectrolytes (CPEs) as ETL was introduced to improve the carrier balance as well as the compatibility with the metal cathode. The NPL-LEDs with structure of ITO/PEDOT:PSS/PVK/NPLs/CPE/Ag emitted red light at 658 nm with much improved EQE of 5.73% than that of 0.63% reported previously [4]. The NPL-LEDs also showed high 1540 cd/m^2 luminance together with FWHM of 25 nm. The CIE color coordinates were measured as (0.71, 0.29), corresponding to 98% saturated red light. All these characteristics demonstrated the potential capabilities of NPL-LEDs for display applications.

In 2019, Kelestemur et al. reported LEDs using CdSe/CdS/CdZnS core/shell NPLs as the emitter [94]. The diagram of band alignment is shown in Figure 6(a) with PVK and ZnO NPs used as HTL and ETL, respectively. Different from the previous report [4], they used CdSe NPLs with a graded shell consisting of a CdS buffer layer and a $\text{Cd}_x\text{Zn}_{1-x}\text{S}$ gradient layer (Figure 6(b)). The improved optical properties of the graded shell NPLs showed low amplified spontaneous emission as low as $\sim 40 \mu\text{J/cm}^2$ (Figure 6(c)). The graded shell NPL-LEDs with EL peak at 650 nm (Figure 6(d)) exhibited much higher EQE (9.92% vs. 1.80%) and maximum luminance (46000 cd m^{-2} vs. 10650 cd m^{-2}) compared with conventional CdSe/CdS NPLs (Figure 6(e)). This was attributed to the imbalanced charge injection caused by the similar conduction band edges of CdS and ZnO. Thus, the addition of a $\text{Cd}_x\text{Zn}_{1-x}\text{S}$ shell added an additional barrier for electron injection and provided proper surface passivation. The devices exhibited a steep increase in current density and luminance behavior above the turn-on voltage of $\sim 2.3 \text{ V}$.

Recently, Liu et al. reported NPL-LEDs based on CdSe/CdZnS core/shell NPLs [95]. The device architecture was ITO/ZnO/NPLs/CBP/ MoO_3 /Al (Figure 7(a)). Several strategies were used to enhance the device performance. First, the hot-injection shell (HIS) growth of NPLs resulted in a smooth film surface, reduced nonradiative recombination, and enhanced stability. The PLQY was reaching 95% in solution and 87% in film even after rigorous cleaning (Figure 7(c)). In comparison, PLQY of core/shell NPLs grown by c-ALD was usually less than 11%, leading to poor device efficiency (Figure 7(d)). Second, the maximum EQE showed strong dependence on the AR where the EQEs of LEDs consisting of NPLs with AR of 3.2, 1.6, and 1.0 (Figure 7(b)) were 5.66%, 8.65%, and 11.19%, respectively. This can be attributed to the low root mean square (RMS) roughness of low AR (2.43 nm) NPL films compared to that of high AR (5.74 nm) and mid-AR (3.44 nm) NPL films. As a result, the leakage currents were remarkably reduced in low AR NPL-LEDs. Finally, CPB can easily crystallize due to

its low glass transition temperature (T_g) of 62°C , leading to morphological instability of LEDs. To overcome this problem, HTL materials with better stability, $\text{N,N}'$ -di-(1-naphthyl)- $\text{N,N}'$ -diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB)/4,4',4''-tris(carbazol-9-yl)-triphenylamine (TCTA), were used instead of CPB since TCTA has a HOMO (5.7 eV) [96] located between the HOMO of NPB (5.3 eV) [97] and the VBM of CdSe/CdZnS. The NPL-LEDs based on HIS NPLs exhibited very high EQE of 19.23% (Figure 7(e)) and a maximum luminance of 23490 cd m^{-2} . This is the first time that the EQE of NPL-LED can be comparable with that of OLEDs and PeLEDs. It is concluded that several conditions need to be satisfied for an ideal NPL-LED. First, the PLQY of NPLs needs to be as high as possible or close to 100%. This can be only achieved by the core/shell structure synthesized by the hot-injection method to passivate nonradiative recombination pathways. Second, the ETL and HTL materials need to be optimized for the best charge injection efficiency and balance. Third, since the major advantage of NPL-LEDs over QLEDs is the in-plane TDM that can break the EQE limit of 20%, successful assembly of NPL films containing only in-plane TDM is critical to realizing the high EQE. We will discuss recent progress towards oriented NPL films in Section 4.3.

Besides red NPL-LEDs studied by most reports, Zhang et al. reported a green LED based on colloidal CdSe/CdS core/shell NPLs with a narrow PL at 556 nm and FWHM of 12 nm [98]. They synthesized CdSe core NPLs first and found that the PL spectra shifted to longer wavelengths (from 361 to 556 nm) with the increase in reaction time, while the PL FWHM decreased from 30 nm to 12 nm at the same time. In order to obtain high-purity green emission, they used the CdSe core with reaction time of 10 min and PL at 554 nm to grow CdSe/CdS core/shell NPLs. The growth of shells was along with the edge of CdSe NPLs, which was indicated by the uniform thicknesses of $\sim 2.2 \text{ nm}$ for both core and core/shell NPLs. Therefore, it is possible that these NPLs were core/crown structure instead of core/shell. The device used a conventional structure of ITO/PEDOT:PSS/TFB/NPL/ZnO/Al (Figure 6(e)). The NPL-LEDs not only exhibited several hundred times increase in brightness but also possessed supernarrow EL with FWHM of 14 nm. The ZnO NP ETL has electron affinity of $\sim 4.3 \text{ eV}$ and ionization potential of $\sim 7.6 \text{ eV}$, providing both the efficient electron injection channel and barriers confining holes within NPLs (Figure 6(f)). The valence band offset at the NPL/ZnO NP interface led to improved charge recombination efficiency. These devices exhibited maximum luminance efficiency of 12.5 cd A^{-1} , corresponding to the highest EQE of 5% at the brightness of $\sim 2000 \text{ cd m}^{-2}$. Although the efficiency is lower than those of previous reports which utilized sphere-core/shell QDs, the NPL-based green LEDs still have great potential for green LEDs.

4.1.3. Core/Crown CdSe NPL-LEDs. The heterostructure NPLs can be classified as type I or type II which depends on the band alignment between the core NPLs and the shell or crown [99–100]. In type I NPLs, both electron and hole wave functions are confined in the core as a result of the

FIGURE 6: (a) Energy band diagram of the optimized device structure for graded shell CdS/Cd_xZn_{1-x}S NPL-LEDs. (b) Schematic of synthesis of graded shell NPLs. (c) Optical gain characteristics of CdSe/CdS/Cd_xZn_{1-x}S NPLs showing a low ASE threshold of ~40 μJ/cm². (d) EL spectra of NPL-LEDs. Inset: the photo of the fabricated LED in operation. (e) EQE vs. current density representation of NPL-LEDs. Adapted with permission from reference [94]. Copyright 2019 American Chemical Society

wider band gap of the shell or crown than that of the core. In contrast, the electron and hole wave functions are spatially separated in type II NPLs. Since the first type II NPLs were synthesized in 2015 [101], type II NPLs have shown great potential to develop LEDs with tunable emission colors ranging from the visible to near infrared (NIR). Additionally, type II NPLs can maintain high PLQY and reduced overlap between absorption and emission spectra which is important for achieving high EQE.

In 2018, Liu et al. studied LEDs based on CdSe/CdSe_{0.8}Te_{0.2} core/crown type II NPLs [38]. The PL spectrum of the type II CdSe/CdSe_{0.8}Te_{0.2} core/crown NPLs showed a red emission peak at ~599 nm. The device used inverted structure of ITO/ZnO/NPLs/TCTA/TPD/MoO₃/Al. The high EQE of 3.57% was ensured by the high PLQY (85%) of core/crown NPLs. More importantly, the dual HTL of TCTA/TPD worked much better than the conventional CBP or TPD. This is attributed to the following: (i) TCTA

FIGURE 7: (a) Device architecture of the NPL-LEDs. (b) HR-TEM images of high, mid, and low AR NPLs. (c) Evolution of the PLQY of core/shell NPLs. (d) EQE of NPL-LEDs with varying shell composition. (e) EQE histogram of the NPL-LEDs. Adapted with permission from reference [95]. Copyright 2019 Wiley-VCH GmbH.

is hole-dominating and has a higher LUMO to confine the electrons. Hence, more excitons can form when more holes can meet electrons; (ii) the stepwise HOMO of TCTA/TPD reduces the hole barrier and injects more holes into the NPLs; (iii) TCTA/TPD-based NPL-LEDs also have better charge balance, with the high electron injection efficiency owing to the high electron mobility and LUMO barrier of ZnO [7]; and (iv) the charge neutrality for NPLs can be maintained while keeping good emissive properties due to the dual-hole transport layer [102]. In addition, the inverted structure is more favorable for display applications as the cathode can be directly connected to the low-cost *n*-type metal-oxide or amorphous-silicon thin film transistors [103]. In summary, the core/crown NPL-LEDs exhibited an extremely low turn-on voltage of 1.9 V, a high EQE of 3.57%, and a PE of 9.44 lm W⁻¹. Finally, this work demonstrated that it is necessary to focus more on the optimization of device structure.

Recently, Wen et al. reported another core/crown CdSe/CdS NPL-LED [105]. This device exhibited an electroluminescence (EL) at 521 nm with a FWHM of 15 nm. The device used inverted architecture of ITO/ZnO/PVP/NPLs/CBP/MoO₃/Al due to the better solvent compatibility of ZnO than HTL materials. The thin PVP layer was introduced to prevent the direct contact between the ETL and the HTL caused by the cracks of the NPL layer. Moreover, the PVP can also slower electron transport and improve the charge balance. As a result, these devices exhibited a maximum

luminance efficiency of 4684 cd m⁻², an EQE of 0.416 %, and a current efficiency (CE) of 1.46 cd A⁻¹, which were 7-fold brightness and 6-fold EQE of those from NPL-LEDs without a PVP layer. This work demonstrated that adding a PVP layer is a great way to enhance performance of NPL-LEDs. The EQE of core/crown NPL-LEDs can be further improved through reducing the stacking NPLs (edge-up orientation). Wen et al. showed that the increasing polar solvent (ethanol) can effectively reduce the staking between NPLs, thus reducing nonradiative energy transfer (NRET) between NPLs (Figure 8(a)) [104]. The NPL-LEDs with structure of ITO/ZnMgO/NPLs/CBP/MoO₃/Al illuminated pure green light at 521.5 nm with FWHM of 15 nm, corresponding to CIE coordinates of (0.108, 0.800). The suppressed NRET resulted in an EQE of 2.16% and a maximum luminance of 22500 cd m⁻², both of which are several times of those from the previous report.

4.1.4. Alloyed Core-Only NPLs for LEDs. One limitation of NPLs is the discrete wavelengths that depend strictly on atomic layer thickness [106–107]. To overcome this problem, Fan et al. used alloyed CdSe NPL with CdS to finely tune the emission spectrum while still leveraging atomic-scale thickness control [7]. They used 6-ML CdSe NPLs alloyed with CdS of which the emissions could cover from green to blue (513–481 nm). Different HTL materials, CBP, NPB, and 4,4'-cyclohexylidenebis(N,N-bis (4-methylphenyl)-benzenamine) (TAPC), were compared, and CBP/

FIGURE 8: (a) Schematic of reducing the stacking NPLs by solvent optimization. (b) A photograph of NPL-LED emitting pure green light. (c) CIT coordinates of the NPL-LEDs. Adapted with permission from reference [104]. Copyright 2021 Wiley-VCH GmbH. (d) The CdSe-CdS alloyed NPL-LED under zero bias and forward bias near turn-on voltage (2 V), modeled using SCAPS. (e) Energy band diagram of NPL-LEDs. (f) Tunable EL spectra from NPL-LEDs. Adapted with permission from reference [7]. Copyright 2015 American Chemical Society.

MoO₃ bilayer HTL was selected for the best performance. The inverted device architecture was ITO/ZnO (50 nm)/NPLs (70 nm)/CBP (65 nm)/MoO₃ (10 nm)/Ag (100 nm) (Figure 8(d)). The energy band diagram is shown in Figure 8(e). As a result, the NPL-LED showed EL wavelength from 491 nm to 513 nm with the narrowest EL FWHM of 12.5 nm (Figure 8(f)), a peak brightness of $\sim 90 \text{ cd m}^{-2}$, and a subband gap turn-on voltage of 2.1 V. This work stated that more efforts were needed to develop the core-only NPLs.

4.2. Perovskite Nanoplatelet-Based LEDs. Metal halide perovskites have attracted a lot of interests for a variety of optoelectronic applications, such as lasers, photovoltaics, and LEDs [108–112]. Particularly, perovskite-based LEDs have drawn lots of attention because of their high PLQY [113–114], narrow emission bandwidth, and tunable EL [115–116]. Since the development of LEDs based on CH₃NH₃PbBr₃ perovskite in 2014 [117], many efforts have been made for this new type of LEDs [118–123]. Compared with other materials like organic semiconductors and inorganic quantum dots, perovskites have many advantages, for example, low material cost, easy synthesis, and high color purity.

4.2.1. Hybrid Organic-Inorganic Perovskite NPL-LEDs. Organic-inorganic hybrid perovskites, particularly the three-dimensional (3D) perovskites, have emerged as an excellent

class of semiconductors due to their high PLQY, long carrier diffusion length, and long carrier lifetime [124–126]. These properties have enabled high-performance LEDs, lasers, solar cells, and many other optoelectronic devices [108, 127–129]. Due to the limited band gap, it has been difficult to achieve blue EL from LEDs based on 3D perovskites. 2D perovskites, on the other hand, have larger band gaps because of the quantum confinement. Liang et al. achieved the first violet LED using 2D perovskite (C₆H₅CH₂CH₂NH³⁺, PEA)₂PbBr₄ thin film [130]. They applied a solvent vapor annealing technique to prepare high-quality micrometer-lateral (PEA)₂PbBr₄ nanoplatelets. The device structure was ITO/PEDOT:PSS (30 nm)/(PEA)₂PbBr₄/1,3,5-tris(1-phenyl-1H-benzo[d]imidazol-2-yl) benzene (TPBi, 35 nm)/Ca (25 nm)/Al (100 nm) (Figure 9(a)). The LED displayed an EL emission peak at 410 nm with FWHM of 14 nm and an EQE of 0.002% (Figure 9(b)).

Perovskite NPLs with high crystallinity and high PLQY are critical for NPL-LEDs [130]. Ling et al. synthesized CH₃NH₃PbBr₃ (MAPbBr₃) NPLs with PLQY up to 85% and fabricated bright perovskite NPL-LEDs [120]. The device structure was ITO/PEDOT:PSS/PVK:2-(4-biphenyl)-5-phenyl-1,3,4-oxadiazole (PBD)/NPLs/bathocuproine (BCP, 50 nm)/LiF (1 nm)/Al (150 nm) (Figure 9(c)). The NPL-LEDs exhibited maximum luminance of $10,590 \text{ cd m}^{-2}$ (530 nm), PE of 1.0 lm W^{-1} , and EQE of 0.48%. Besides high-quality NPLs, bipolar PVK:PBD in the emitting layer

FIGURE 9: (a) The SEM for LEDs (scale bar: 200 nm). (b) Normalized luminescence of the LED using $(PEA)_2PbBr_4$ NPLs achieved from dimethylformamide vapor annealing at 6 V. EL and PL emission peaks are peaked at 410 and 407 nm, respectively. Inset: a picture for violet light from the $(PEA)_2PbBr_4$ LED; adapted with permission from reference [130]. Copyright 2016 American Chemical Society. (c) Device architecture and the energy levels of the LED. (d) Current-voltage and luminance-voltage properties of the control LED (neat perovskites without PVK:PBD) and LEDs using the PVK:PBD (ratio of 70:30, 64:36, and 58:42) layer. (e) EQE-current density of LEDs. Adapted with permission from reference [120]. Copyright 2015 Wiley-VCH GmbH.

also improved charge balance (Figures 9(d) and 9(e)). In comparison, the device without the PVK:PBD only showed EQE of 0.038% and luminance of 1113 cd m⁻². Additionally, the MAPbBr₃ NPLs also showed good stability in air for more than a week.

Similarly, Kumar et al. adopted the MAPbBr₃ NPLs as emitters for LEDs [131]. Two strategies were adopted to achieve blue emission from MAPbBr₃ NPLs. First, the thickness was precisely controlled by cosurfactant colloidal chemistry to yield a high degree of blue shift. Second, the wide-band-gap and low-*k* organic hosts (e.g., CBP) as the barrier materials formed perovskite dielectric quantum wells which can enable efficient radiative recombination by the significantly increased exciton binding energy. These results are useful for perovskite LEDs to cover the entire visible spectrum.

Like MAPbBr₃, formamidinium lead bromide (FAPbBr₃) NPL is also an efficient emitter for LED. Kumar et al. reported a pure green LED using FAPbBr₃ NPLs with thickness of 7~10 unit cells [132]. The quantum-confined FAPbBr₃ NPLs exhibited high exciton-binding energy of 162 meV through the dielectric quantum well engineering, leading to high PLQY of 92% in a thin film. By optimizing both the HTLs and ETLs, the best structure of ITO/PEDOT:PSS/poly-TPD/PMMA:FAPbBr₃/3TPYMB/LiF/Al was

selected. As a result, the optimized device showed a maximum current efficiency of 13.02 cd A⁻¹ at 529 nm with a FWHM of 22.8 nm. Moreover, the LED reached CIE 1931 color coordinates of (0.168, 0.773) and a wide color gamut covering 97% and 99% of the Rec. 2020 standard in the CIE 1931 and the CIE 1976 color space. Compared with the LED without PMMA, the efficiency of the device based on the 2D FAPbBr₃-PMMA complex improved by 3 times, which can be attributed to the dielectric confinement effect and the formation of a smoother EML. Moreover, efficient exciton recombination and balanced charge carriers also promoted the device performance due to the following: (i) the deep LUMO of 3TPYMB (-3.3 eV) and the shallow HOMO of poly-TPD (-5.4 eV) can enable the cascade carrier injection; (ii) carrier mobilities in 3TPYMB and poly-TPD are comparable; (iii) the high LUMO of poly-TPD (-2.0 eV) and the low HOMO of 3TPYMB (-6.8 eV) can form an effective carrier confinement.

4.2.2. All-Inorganic Perovskite NPL-LEDs. Hybrid organic-inorganic perovskites have been well demonstrated to develop high-efficiency LEDs. However, due to the small formation energy, the organic cations like $CH_3NH_3^+$ are sensitive to moisture and can easily decompose. On the contrary,

FIGURE 10: (a) EQE and luminance as a function of current density. (b) EL spectra measured at various voltages. Inset: a picture of LED at 10 mA cm⁻²; adapted with permission from reference [141]. Copyright 2017 American Chemical Society. (c) EL spectrum at an applied bias of 4 V. Inset shows a photograph of a 56 mm² device. (d) Current density-luminance-voltage characteristics and (e) corresponding EQE-luminance curve of a device. (f) Histogram of peak EQEs for 32 devices from five batches. Adapted with permission from reference [143]. Copyright 2019 American Chemical Society.

all-inorganic perovskites like CsPbBr₃ may overcome these stability issues while maintaining high PLQY and narrow emission [133–135]. LEDs based on inorganic perovskites have attracted a lot of interest in the past few years [116, 136–139]. Zhang et al. used an inorganic perovskite composite of CsPbBr₃ NCs and quasi-2D CsPb₂Br₅ NPs as emitters and demonstrated LEDs exhibiting a maximum luminance of 3853 cd m⁻², an EQE of 2.21%, and a current efficiency of 8.98 cd A⁻¹, peaking at 527 nm with a FWHM of ≈24 nm [140]. The device architecture was ITO/PEDOT/CsPbBr₃-CsPb₂Br₅/TPBi/LiF/Al. Smaller-sized CsPb₂Br₅ NPs attached on the surface of cubic CsPbBr₃ NCs can improve current efficiency by reducing exciton diffusion length and enhance emission lifetime by reducing nonradiative energy transfer to trap states. This dual-phase all-inorganic CsPb₂Br₅-CsPbBr₃ composite nanocrystal provided a new route of perovskite-based LEDs.

Qin et al. used pure quasi-2D CsPb₂Br₅ NPLs as emitters for LEDs [141]. By centrifugal coating technology, CsPb₂Br₅ NPL films with a PLQY of ~35% were produced. The device architecture was ITO (100 nm)/PEDOT:PSS (30 nm)/CsPb₂Br₅ (30 nm)/TPBi (40 nm)/LiF (0.8 nm)/Al (100 nm). The NPL-LEDs emitted at 520 nm with a FWHM of 20 nm (Figure 10(b)) and had a turn-on voltage of 3.8 V, a maximum luminance of 7317 cd m⁻² (Figure 10(a)), a maximum EQE of 1.1%, and CIE coordinates of (0.08, 0.75). Furthermore, CsPb₂I₅ NPLs were acquired by halogen exchange,

and the CsPb₂I₅ LEDs emitted at 685 nm and had an optical band gap of 1.75 eV, a lower PLQY of ~15%, and a maximum EQE of 0.14% at 20 mA cm⁻². Similar to this work, Han et al. also studied CsPb₂Br₅ NPLs as emitters for LEDs [142]. CsPb₂Br₅ NPLs with tunable emission wavelengths from blue (467 nm) to bright green (518 nm) were achieved by changing the temperature from 100°C to 140°C. They found that CsPb₂Br₅ NPLs synthesized at 140°C were capable of emitting narrow-band green light (FWHM ~18 nm). Under an operation current of 10 mA, the green LEDs showed a luminous efficiency of 34.49 lm/W.

2D CsPbX₃ NPLs (X = I, Br, and Cl) have drawn enormous attention since Bekenstein et al. reported CsPbX₃ NPLs in 2015 [61]. 2D CsPbX₃ NPLs are promising materials for optoelectronic applications due to their precisely tunable thickness [131]. Si et al. reported 2D layered PBA₂ (CsPbBr₃)_{n-1}PbBr₄ perovskites (*n* = 12–16) where the CsPbBr₃ NPLs were passivated by bulky phenylbutylammonium (PBA) cations [143]. By controlling the *n* values, the thickness-controlled quantum well (TCQW) CsPbBr₃ films were obtained with quantum confinement effects. The device architecture was ITO/NiO/TFB/PVK/CsPbX₃/Ca/Al. Due to smooth surface features, low trap densities, narrow emission line widths (~82 meV, Figure 10(c)), and high PLQY of TCQW CsPbBr₃ films, the NPL-LEDs exhibited high EQEs of up to 10.4%, maximum luminance of 14000 cd m⁻² at 8 V, and high color purity (Figures 10(d)–10(f)). Besides, this

device also showed good stability because no significant decrease in device efficiency was observed after 30 days in a glovebox. Finally, CsPbI₃ NPL-LEDs were obtained by the same approach. The EL spectrum of CsPbI₃ NPL-LEDs displayed an emission peak at 683 nm with a FWHM of 90 meV, corresponding to CIE color coordinates of (0.72, 0.28) and a peak EQE of 7.3%.

Tian et al. used quasi-2D (BA)₂(Cs)_{n-1}[Pb_nI_{3n+1}]/poly(ethylene oxide) (PEO) composite thin films as emitters [144]. The device architecture was ITO/PEDOT:PSS (40 nm)/poly-TPD (20 nm)/EL (50 nm)/TPBi (40 nm)/LiF (1 nm)/Al (100 nm). By changing the molar ratios of organic salt (BAI) to inorganic salts (CsI and PbI₂), the quasi-2D films with emission at 638–690 nm were obtained. Due to the surface passivation and small emitting crystals formed in the composite thin films which significantly reduced the nonradiative recombinations, the NPL-LED with emission peak at 680 nm exhibited a brightness of 1392 cd m⁻², an EQE of 6.23%, and great stability.

Besides green emitters or red emitters, Yang et al. reported ultrathin CsPbBr₃ NPLs for blue LED [145]. The thickness of CsPbBr₃ NPLs can be precisely tuned in the monolayer level by controlling the reaction kinetics. Moreover, the edge length can also be varied broadly through modifying the reaction time. The device architecture was ITO/PEDOT:PSS/poly-TPD/CsPbBr₃/TPBi/LiF/Al. The device showed a maximum luminance of 25 cd/m² and an EQE of ~0.1%. This was the first time that pure CsPbBr₃ NPLs were used as the blue LED, and more efforts are still required to further improve the performance.

Hoye et al. demonstrated that the key limiting factors of blue-emitting NPL perovskite LEDs were the large barriers for hole injection and substantial nonradiative decay at the NPL/HTL interface [146]. Both issues arose in part due to the deep ionization potentials (≥ 6.5 eV) of NPLs, leading to the more difficult hole injection than electron injection and resulting in higher electron current density than hole current density. To overcome these issues, they used a poly(triarylamine) interlayer (poly-TPD for sky blue NPLs; TFB for blue NPLs) between the PEDOT:PSS layer and emitter layer to reduce the nonradiative loss and increase hole injection efficiency. As a result, the EQE of the blue (464 nm) and sky blue (489 nm) LEDs were increased to 0.3% and 0.55% which were higher than previous reports.

For further development, Zhang et al. used the surface ligand engineering strategy to enhance the PLQY and stability of perovskite NPLs [147]. The device architecture was ITO/PEDOT:PSS/(poly-TPD)/CsPbBr₃/TPBi/LiF/Al (Figure 11(a)). By replacing the original long-chain OLA with halide ion-pair ligand (didodecyl dimethyl ammonium bromide (DDAB)), significant enhancement of PLQY and stability was achieved while no significant change was observed in morphology and crystal structure of the NPLs. Due to the halide ion-pair ligand, the CsPbBr₃ NPL films were more uniform with a lower RMS roughness (0.99 nm) compared with NPLs with OLA ligands (1.60 nm). Besides, the hole injection barrier between the poly-TPD and the CsPbBr₃ NPLs was reduced. After the ligand exchange, the CsPbBr₃ NPL-LEDs showed maximum luminance of

41.8 cd/m², peak EQE of 0.56%, and PE of 0.4 lm/W. Since the remaining hole injection barrier (1.5 eV) was still relatively large, they inserted a thin CBP layer as another HTL between the CsPbBr₃ NPLs and the poly-TPD HTL to further improve the charge balance and facilitate the hole injection. Consequently, the peak EQE reached 1.42% and PE reached 1.33 lm/W (Figures 11(c)–11(f)). This work demonstrated that the surface ligand engineering played an important role in improving PLQY and stability of perovskite NPLs and the performance of LEDs.

4.3. Oriented NPL Films for Improved Out-Coupling Efficiency. NPLs are considered promising materials for LEDs mainly because the in-plane TDMs provide new opportunities to increase the out-coupling efficiency and thus break the EQE limit of ~20% for QLEDs [148]. Compared with isotropic NCs (QDs) which are difficult to align their TMDs due to the isotropic shape, the anisotropic NPLs can be assembled into uniform film with unity in-plane TDM distribution; that is, all NPLs are in face-down orientation. Recently, several reports have demonstrated controlled assembly of NPLs with all TDMs aligned in-plane [149–151]. Additionally, NPL-LEDs with oriented NPL films were also proven with EQEs up to 24.2% [152–153]. Gao et al. used liquid-liquid interfacial assembly to control the orientation of CdSe NPLs [149]. The monolayer CdSe NPL film can be assembled by adding the NPLs in hexane dispersion to the surface of diethylene glycol (DEG). Then, the monolayer film can be transferred to a substrate submerged in DEG as the DEG was slowly drained from the bottom of the container. It was found that the amount of additional oleic acid added to the DEG was critical to determining the orientation of NPLs by tuning the interfacial energy between the NPLs and the NPL-DEG interface. However, the insulating additive may affect the charge transport. To overcome this issue, Momper et al. developed an improved method through controlled solvent evaporation [150]. They select acetonitrile instead of DEG for the higher interaction potential between the NPL flat surface and the liquid. In addition, the evaporation rate of the solvent where NPLs were dispersed in was also critical, where fast evaporation either by increasing the temperature or selecting high vapor pressure solvent can result in face-down assembly.

In case of perovskite NPLs, Kumar et al. reported efficient NPL-LEDs based on oriented FA_{0.5}MA_{0.5}PbBr₃ NPLs [152]. The perovskite NPLs were directly assembled on the substrate by spin coating. Owing to the shear-induced ordering during spin coating, the majority of NPLs were in face-down orientation. Comparably, drop casting resulted in random orientation containing both face-down and edge-up orientations. The NPL-LEDs with optimized device structure of ITO/PEDOT:PSS/X-F6-TAPC/NPLs/3TPYMB/LiF/Al exhibited record high EQE of 24.2% covering a wide range of luminance between 30 and 1500 cd⁻². The emission wavelength was at 528 nm with a FWHM of 22 nm. Recently, Cui et al. also reported perovskite NPL-LEDs using oriented inorganic perovskite CsPbBr₃ NPLs [153]. The CsPbBr₃ NPLs were directly grown on the PVK HTL by spin coating the precursor solution followed by annealing at

FIGURE 11: (a) Schematic device architecture of the CsPbBr₃ NPL-based LED. (b) EL spectra of the untreated and treated CsPbBr₃ NPL-based LEDs. Inset: a photograph of a working treated CsPbBr₃ NPL-based LED at a driving voltage of 5 V. (c) Current density-voltage, (d) luminance-voltage, and (e) external quantum efficiency-current density curves of the untreated and treated CsPbBr₃ NPL-based LEDs. (f) External quantum efficiency-current density curve of the treated CsPbBr₃ NPL-based LED with a bilayer HTL. Adapted with permission from reference [147]. Copyright 2019 American Chemical Society.

TABLE 2: Summary of the performance of representative NPL-LEDs.

Chemical composition	λ (nm)	PLQY (%)	EQE (%)	CIE	Ref
CdSe	515	30	0.63	—	4
CdSe/CdS/CdZnS (core/shell)	646	89	9.92	(0.73, 0.29)	94
CdSe/CdZnS (core/shell)	650	95	19.23	(0.715, 0.283)	95
CdSe/CdS (core/shell)	556	85	5	(0.316, 0.651)	98
CdSe/CdSe _{0.8} Te _{0.2} (core/crown)	599	85	3.57	(0.61, 0.38)	38
CdSe/CdS (core/crown)	521	84	0.416	(0.101, 0.806)	105
CdSe-CdS (alloyed)	481-513	—	—	—	7
(PEA) ₂ PbBr ₄ NPL	410	85	0.002	—	130
MAPbBr ₃	530	85	0.48	—	120
FAPbBr ₃	529	92	2.21	(0.168, 0.773)	132
2D CsPb ₂ Br ₅ -3D CsPbBr ₃	527	83	2.21	—	140
CsPb ₂ Br ₅	520	35	1.1	(0.08, 0.75)	141
CsPb ₂ Br ₅	467-518	63	—	(0.25, 0.69)	142
PBA ₂ (CsPbBr ₃) _{n-1} PbBr ₄	514	55	9.7	(0.07, 0.74)	143
CsPbBr ₃	480	—	0.1	—	145
CsPbBr ₃	464	19	0.3	—	146
CsPbBr ₃	489	27	0.55	—	146
CsPbBr ₃	469	69	1.42	—	147
FA _{0.5} MA _{0.5} PbBr ₃	528	—	24.2	—	152
CsPbBr ₃	518	75	23.6	(0.09, 0.78)	153

100°C. The key factor affecting the growth of face-down NPLs was found to be the bulky organic ammonium cations in the precursor solution which was prepared by mixing PBABr, PEABr, CsBr, and PbBr₂ in DMSO. Although not completely understood yet, the formation of face-down NPLs may originate from the van der Waals interactions. It was also found that adding LiBr into the precursor solution can significantly improve the PLQY from ~50% to 75% due to the better surface passivation. The NPL-LEDs adopted the device structure of ITO/NiO_x/TFB/PVK/NPLs/TPBi/LiF/Al which demonstrated a high EQE of 23.6% from a champion device and an average EQE of 21.3% from 36 devices. The emission was at 518 nm with a narrow FWHM of 16 nm, corresponding to CIE color coordinates of (0.09, 0.78). With further development, the EQE of oriented NPL-LEDs is expected to reach the upper limit of 40% very soon.

5. Summary and Outlook

Over the past few years, NPL-LED technology has experienced fast development, especially in the synthesis of NPLs. Due to their unique optoelectronic properties, NPLs have attracted significant attention for various fields, and many efforts have been made to improve the properties including PLQY, FWHM, and stability. For NPL-LEDs, the charge carrier balance is a critical factor affecting the performance. Through modulating potential barriers, charge balance in EMLs can be improved, resulting in high formation probability of excitons, low nonradiative recombination rates, and reduced overflow current of excess carriers. Optimizing

the structure or composition of NPLs, for example, core/shell, core/crown, and alloyed NPLs, can increase PLQY by prohibiting nonradiative recombination. For NPL-LEDs, understanding the interfacial physics is critical to enhance the performance. The exciton quenching at the interface of charge transport layer/NPLs has been greatly suppressed, and the potential energy barrier has been reduced via modifying the interface of NCs films or HTL (e.g., ligand exchange). In virtue of these advances, the EQEs of red NPL-LEDs based on CdSe/ZnCdS NPLs have reached nearly 20%. Although the performance of NPL-LEDs has been significantly enhanced, they still lag behind state-of-the-art spherical QD-LEDs and OLEDs. However, it is believed that NPL-LEDs have the potential to be comparable to their counterparts. Table 2 summarizes the properties and performance of different NPL-LEDs. CdSe-based NPLs have been shown as great EL for red and green LEDs. Due to the intrinsically low band gap of CdSe, it is challenging to achieve blue or deep blue LED from CdSe NPLs. On the other hand, perovskite NPLs, consisting of a large family of perovskite materials, can potentially cover the entire color space required for LEDs. Currently, the research of perovskite NPL-LEDs has been focusing on inorganic CsPbBr₃ due to the simpler synthesis and better stability compared with its organic-inorganic counterparts. However, the perovskite NPL-LEDs still suffer very limited lifetime compared with CdSe NPL-LEDs. Nevertheless, the study of NPL-LEDs only emerged less than a decade ago; thus, many fundamental issues remain unsolved and urge further exploration.

There are a few issues still waiting to be solved before NPL-LEDs can be successfully commercialized. First, although CdSe-

based NPL-LEDs have exhibited higher EQE for red and green emission, the toxicity of Cd strictly limited the use of Cd-containing NCs in practical applications. Thus, the studies of environmentally friendly non-Cd-containing NCs and their LEDs are urgently required. Second, many improvements are left to be done for NPL-LEDs, such as the EQE, PE, and lifetime. These can be achieved by employing NPLs with high PLQY and stability, carefully manipulating charge distribution, and introducing the out-coupling technique [154–155]. In addition, energy barrier and material selection also need to be managed [156–158]. Finally, blue NPL-LEDs currently lag behind in device efficiency and lifetime compared to green and red NPL-LEDs. The longest operation lifetimes of blue NPL-LEDs are far below the minimum requirement for display applications (10000 h), primarily due to the low-quality blue emitting materials with deep-lying VBM, which is not sufficient for efficient hole flow across the device. Hence, new HTL materials with deeper HOMO/VBM level as well as higher mobility are required to realize charge balance and enhance the lifetime and the efficiency of blue NPL-LEDs. Moreover, surface modification can be introduced to reduce the potential energy barrier and improve the performance of NPL-LEDs.

Despite many challenges remaining unsolved at present, with further research and development of the NPL-LEDs, anisotropic nanocrystals are promising materials to be widely applied in display- and light-emitting-related applications in the future.

Conflicts of Interest

The authors declare no conflict of interest regarding the publication of this article.

Authors' Contributions

Yating Guo and Feng Gao contributed equally to this work

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