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Article

YCl₃-substituted CsPbI₃ Perovskite Nanorods for Efficient Red Light-emitting Diodes

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Abstract: Cesium lead iodide (CsPbI₃) perovskite nanocrystals (NCs) are a promising material for red light-emitting diodes (LEDs) due to their excellent color-purity and high luminous efficiency. However, small-sized CsPbI₃ colloidal NCs, such as nanocubes, used in LEDs suffer from confinement effects, negatively impacting their photoluminescence quantum yield (PLQY) and overall efficiency. Here, we introduced YCl₃ into the CsPbI₃ perovskite, which formed anisotropic, one-dimensional (1D) nanorods. This was achieved by taking advantage of the difference in bond energies between Cl⁻ and I⁻ ions, which caused YCl₃ to promote the anisotropic growth of CsPbI₃ NCs. The YCl₃-based nanorods improved the PLQY and storage stability by passivating the defects reducing the nonradiative recombination rates. When the YCl₃-substituted CsPbI₃ nanorods are applied to the emissive layer in LEDs, we achieve an external quantum efficiency of ~3.16% which is 1.86-fold higher than the pristine CsPbI₃ NCs (1.69%) based LED. Notably, the ratio of horizontal transition dipole moments (TDMs) in the anisotropic YCl₃:CsPbI₃ nanorods was determined to be 75%, which is higher than the isotropically oriented TDMs in CsPbI₃ nanocrystals (67%). This increased TDM ratio led to higher light outcoupling efficiency in nanorods-based LEDs. Overall, the results suggest that YCl₃-substituted CsPbI₃ nanorods could be promising for achieving high-performance perovskite LEDs.

Keywords: Anisotropic Growth; YCl₃-substituted Perovskite; 1D Nanorods; Transition Dipole Moments; Outcoupling Efficiency

1. Introduction

Metal halide perovskites (MHPs) have gained significant attention due to their outstanding figures of merit, including remarkable color purity, efficient radiative recombination, tailorable emission wavelengths, and low-cost solution-processability. As a result, they are considered a promising candidate for next-generation display and lighting [1–4]. Among various MHPs, all-inorganic CsPbI₃ nanocrystals (NCs) are crucial for these purposes owing to their outstanding chemical and thermal durability [5]. Recent strategies, such as morphology and interfacial control, architecture engineering, and surface chemistry engineering, have been implemented to improve the practical applications of CsPbI₃ NCs, boosting the external quantum efficiency (EQE) of CsPbI₃ NCs-based light-emitting diodes (LEDs) [6]. Despite the rapid progress, the CsPbI₃ NCs LEDs endure substantial limitations due to the small-sized (0-dimension NCs), attributed to the confinement effect [7]. The defects-enriched surface of colloidal CsPbI₃ NCs, owing to their small size, has a deleterious effect not exclusively limited to photoluminescence quantum yield (PLQY), but also coupled with the low-luminous efficiency of LEDs [8]. Moreover, the large surface area of small-size CsPbI₃ NCs requires excessive native ligands to passivate, which forms an insulation layer to impede the effective carrier transport capability in assembled NCs film are numerous arduous challenges that are correlated with CsPbI₃ NCs based LEDs [2].

Anisotropic one-dimensional (1D) nanorods can overcome the limitations of conventional CsPbI₃ NCs-based LEDs. This is because 1D nanorods combine two-directional quantum confinement effects with a significantly reduced surface trap density, leading to excellent photophysical properties and high aspect ratios [9]. The 1D nanorods possess excellent photophysical

properties and high aspect ratios from their unique surface morphology [10,11]. Additionally, the well-defined morphology of 1D nanorods has the ability to confine the active area of charge carriers and shorten the transmission of carriers [3], making them more conducive to the exploration of efficient LEDs compared with their counterpart nanocrystals [12].

While recent reports have described the synthesis of perovskite nanorods from the water-oil transformation of Cs_4PbBr_6 polyhedrons into nanorods or fragmentation of perovskite nanowires initiated by anion-exchange processes [13,14], these methods result in impure morphologies and defects-enriched surfaces. Therefore, it remains challenging to directly synthesize perovskite nanorods with efficient radiative recombination rates (high PLQY) and well-defined aspect ratios. Importantly, the anisotropic nature of these nanorods' emissive layer is valuable to further increase the outcoupling efficiency by overcoming the photons losses that become trapped through waveguiding and total internal, prized for their high ratios of horizontal transition dipole moments (TDMs), contrary to isotopically oriented nanocubes (nanocrystals) [15–17].

Here, an unconventional approach is introduced based on yttrium (III) chloride hexahydrate ($\text{YCl}_3 \cdot 6\text{H}_2\text{O}$) YCl_3 -substituted CsPbI_3 nanorods. The YCl_3 -substituted CsPbI_3 nanorods decrease the lattice parameter of CsPbI_3 NCs owing to partial substitution of Pb^{2+} and I^- with bigger radius by smaller Cl^- ions. The YCl_3 passivates surface traps and regulates the net recombination rates, significantly improving PLQY. Importantly, the environmental durability of the YCl_3 -substituted CsPbI_3 NRs improved significantly, losing 28% PLQY after being stored for 45 days under ambient conditions. Notably, the YCl_3 : CsPbI_3 -nanorod-based LEDs demonstrated a peak EQE of 3.16%, 1.86-fold higher than the control CsPbI_3 NCs-based device (1.69%). Importantly, the ratio of horizontal transition dipole moments of the anisotropic YCl_3 : CsPbI_3 nanorods is determined to be 75%, which is higher than that of the isotopically oriented TDMs in CsPbI_3 nanocrystals (67%), resulting in higher light outcoupling efficiency. Our finding suggests that anisotropic nanorods have a promising prospect in light-emitting devices.

2. Materials and Methods

2.1. Synthesis of unsubstituted and YCl_3 -substituted CsPbI_3 NCs

The previously reported hot-injection method was employed to synthesize the unsubstituted and YCl_3 -substituted CsPbI_3 NCs [18,19].

2.1.1. Cesium Oleate Preparation

Firstly, Cs_2CO_3 (2.49 mmol), OLA (2.5 mL), and 30 mL ODE were degassed and dried at 120 °C for 1 h. Subsequently, the temperature was raised to 150 °C until a clear solution was obtained under N_2 flow.

2.1.2. Synthesis of CsPbI_3 NCs

PbI_2 (0.174 g, 0.376 mmol) and ODE (10 mL) were loaded into a 50 mL three-neck flask, degassed, and dried under vacuum for 1 h at 120 °C. Then, OA (1 mL), and OLA (1 mL) preheated to 70 °C were injected under the protection of N_2 . After the solution became clear, the temperature was raised to 170 °C, and 0.8–1 mL Cs-oleate (pre-heated to >100 °C) was swiftly injected, which was quenched by immersing the flask in an ice-water bath (5 sec later).

2.1.3. Synthesis of YCl_3 : CsPbI_3 NRs

ODE (10 mL), OA (1 mL), and $\text{YCl}_3 \cdot 6\text{H}_2\text{O}$ (0.184 mmol) were loaded into a three-neck flask and adequately dissolved at 150 °C for 1 h under N_2 flow. Then, PbI_2 (0.174 g, 0.376 mmol) and OLA were added, and the temperature was raised to 170 °C (30 min later). Then, as-prepared Cs-oleate was quickly injected into the mixture, and 60–65 s later, the reaction was quenched by immersing the flask in an ice-water bath [12].

2.1.4. Purification

The unsubstituted and YCl_3 -substituted CsPbI_3 NCs were separated by centrifugation at 5000 rpm for 10 min to remove the ODE and unreacted ligands. The precipitate was dispersed in hexane/toluene, and then anti-solvent methyl acetate was mixed, followed by centrifugation at 10000 rpm for 10 min. The resultant mixture was dispersed in 10 mL hexane/toluene and stored in the refrigerator. After 24 h, the supernatant required colloidal ink for LEDs application.

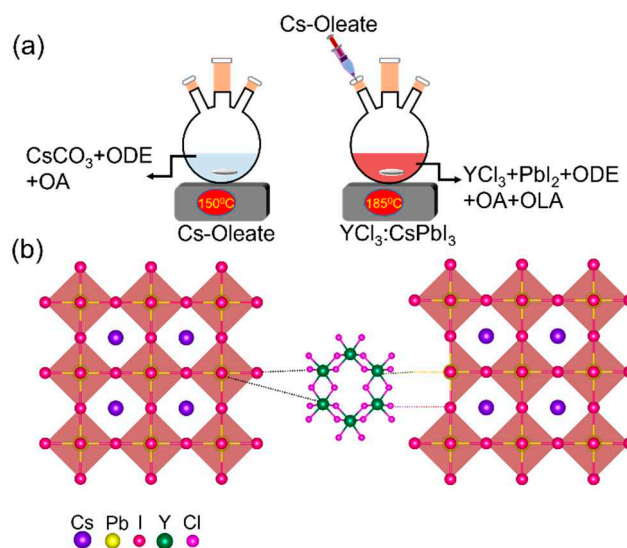
2.2. LED Fabrication

150-nm-thick Indium tin oxide (ITO) patterned glass substrates were cleaned by sequential ultrasonication in acetone and isopropanol for 15 min each. Further, the UV-ozone treatment was carried out for 15 min to improve hydrophilicity before drying with N_2 flow. PEDOT:PSS (filtered with 0.45 μm PVDF filter) was then spin-coated for 1 min at 4000 rpm, which was then annealed at 140 $^\circ\text{C}$ for 30 min. All the substrates were transferred into the N_2 glove box. The hole-transporting Poly-TPD (4 mg/mL in CB) layer was spin-coating onto PEDOT:PSS films at 4000 rpm and baked at 120 $^\circ\text{C}$ for 15–20 min. The perovskite emissive layer was spin-coated at a speed of 2000 rpm for 45 s. Then, 1,3,5-tris(1-phenyl-1H-benzimidazol-2-yl)benzene (TPBi, 40 nm), LiF (1 nm), and Al (130 nm) were sequentially deposited by thermal evaporation in a vacuum deposition chamber. A glass lid encapsulated all the devices with a UV-curable resin in an N_2 -filled glovebox. The optoelectronic properties of LEDs were analyzed using a semiconductor parameter analyzer (Keithley 237) connected with a spectrophotometer (Photo Research PR-670). UV-Vis-IR absorption spectrum of the unsubstituted and YCl_3 -substituted CsPbI_3 NCs film was tested using a PerkinElmer LAMBDA-900 spectrophotometer. PLQY was measured by Quantaaurus-QY Absolute PL quantum yield spectrometer (Hamamatsu, C11347-11). The structure of the unsubstituted and YCl_3 -substituted CsPbI_3 NCs thin film was analyzed by an X-ray diffractometer (X'Pert-PRO MRD, Phillips). The shape of the unsubstituted and YCl_3 -substituted CsPbI_3 NCs was confirmed using a field-emission transmission electron microscope (FE-TEM, JEM-2100F) and the cross-sectional TEM images of the LED were obtained from a Cs-corrected TEM (JEM-ARM 200F, JEOL) installed in the Center for University-wide Research Facilities (CURF) at Jeonbuk National University. Chemical analysis was conducted by X-ray Photoelectron Spectrometer (XPS).

3. Results and Discussion

The control (unsubstituted) and YCl_3 -substituted CsPbI_3 NCs were synthesized following the two-step hot-injection method [20]. The YCl_3 -doped CsPbI_3 NRs were realized by adding 0.184 mmol of $\text{YCl}_3 \cdot 6\text{H}_2\text{O}$ into perovskite medium followed by the injection of Cs-oleate precursor (**Scheme 1a**). Unlike the previous reports that aimed at metal chlorides that used either the same or adjacent chloride element in the pristine perovskite NCs [14,21]. This study examines how metal chloride can modulate the shape and optoelectronic properties of CsPbI_3 NCs. Besides changing the surface defect of perovskite NCs, the YCl_3 -doping has an additional effect on inducing the anisotropic growth of the crystals. The intention doping of YCl_3 was carried out in the CsPbI_3 NCs reaction medium; the distinctive chloride (Cl^-) was not adjacent to the iodide (I^-) of the CsPbI_3 NCs. The presence of chloride on the surface of perovskite nanocrystals (NCs) and the varied bond energies between Cl^- ions and I^- ions are responsible for the anisotropic growth of perovskite NCs. (**Scheme 1b**). The transmission electron microscopy (TEM) analyses provide evidence to support this conjecture, as shown in **Figure 1**. The TEM and high resolution (HR-TEM) morphology of the control and YCl_3 -substituted CsPbI_3 NCs revealed by TEM (Figure 1). The control CsPbI_3 NCs contain monodisperse and regular cubic shape (Figure 1a). The HR-TEM images show a high crystallinity and lattice spacing of 6.2 \AA of the CsPbI_3 NCs, corresponding to the (100) plane of cubic perovskite (Figure 1b-c) [22,23]. The average particle size of the control CsPbI_3 NCs is determined to be ~ 10.05 nm (Figure 1d). Remarkably, the 0.184 mmol YCl_3 -substituted CsPbI_3 NCs show the one-dimensional nanorods (NRs) (Figure 1e-g). The HR-TEM image of YCl_3 -substituted CsPbI_3 NRs displays a lattice spacing of 4.5 \AA , corresponding

to the (110) plane of perovskite (Figure 1g) [12]. The aspect ratio of the YCl_3 -substituted CsPbI_3 NCs is ~ 2.3 , and the average sizes of length and diameter are 18.5 and 8.2 nm, respectively.



Scheme 1. (a) Schematic illustration of hot-injection method of $\text{YCl}_3\text{:CsPbI}_3$ NRs perovskite. (b) The intentional doping of YCl_3 in the perovskite reaction medium. The presence of the surface chloride initiates the unit cell's surface energy imbalance, leading to the anisotropic growth of CsPbI_3 NCs.

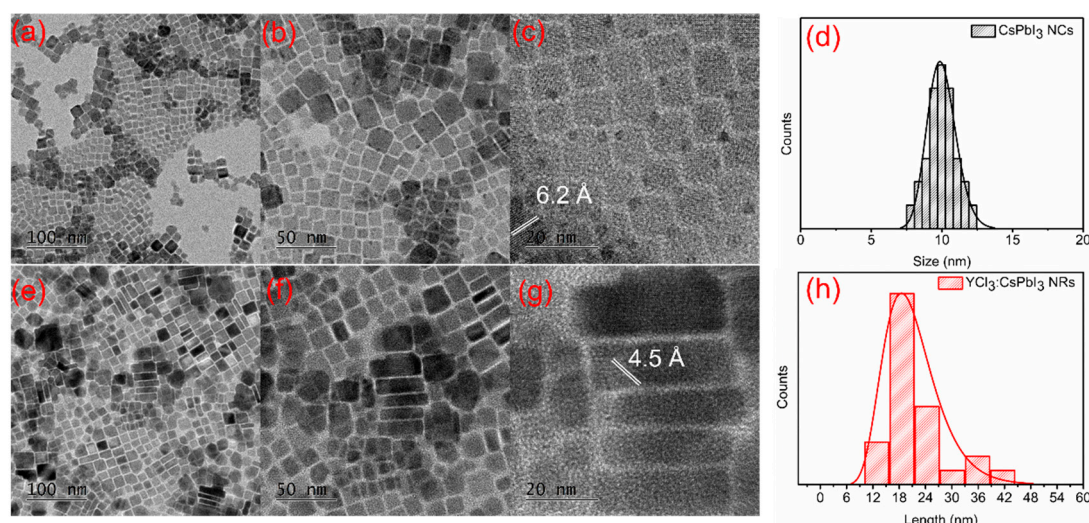


Figure 1. (a) TEM image, and (b-c) high-resolution TEM (HR-TEM) images of CsPbI_3 NCs. (d) The corresponding size distribution histograms. (e) TEM image, and (f-g) HR-TEM images of YCl_3 -substituted CsPbI_3 NRs. (h) The corresponding size distribution histograms.

The X-ray diffraction (XRD) patterns were conducted to ascertain the crystal structure of CsPbI_3 NCs and $\text{YCl}_3\text{:CsPbI}_3$ NRs, as shown in Figure 2. Both the CsPbI_3 NCs and $\text{YCl}_3\text{:CsPbI}_3$ NRs adhere to the reference pattern of the bulk cubic CsPbI_3 perovskite (PDF#98-018-1288), and the diffraction peaks appear at 14.02° , 20.03° , 28.407° , 31.88° , 35.45° , 41.05° , and 51.63° are corresponding to cubic planes of (100), (110), (200), (210), (211), (220) and (300), respectively. The diffraction peaks of (100) and (200) planes shift toward higher angles attributed to the decrease of lattice parameters of $\text{YCl}_3\text{:CsPbI}_3$ NRs stemmed from the partial substitution Pb^{2+} and I^- with bigger radius by the smaller Y^{3+} and Cl^- ions, respectively [12,24]. The scanning electron microscopy (SEM) images of the unsubstituted and YCl_3 -substituted CsPbI_3 NCs are illustrated in Figure 2b-c. Contrary to perovskite NCs, the SEM image of

(YCl₃-substituted CsPbI₃) nanorods are homogeneously distributed on a glass substrate, which indicates that the nanorod film layer has a good foundation for electroluminescent.

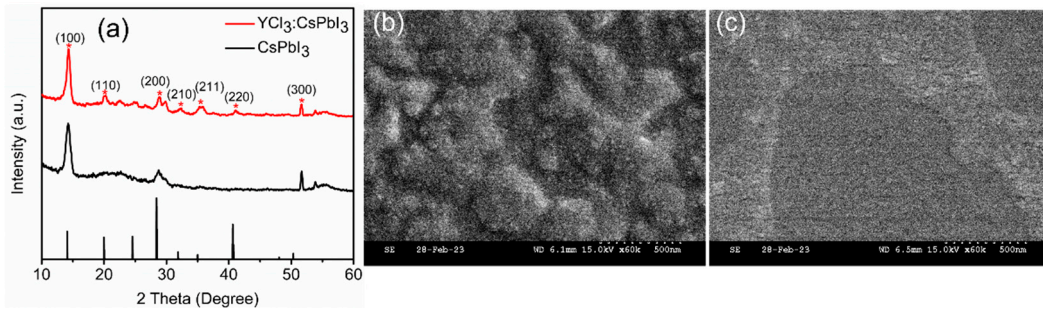


Figure 2. (a) TEM image, and (b-c) high-resolution TEM (HR-TEM) images of CsPbI₃ NCs. (d) The corresponding size distribution histograms. (e) TEM image, and (f-g) HR-TEM images of YCl₃-substituted CsPbI₃ NRs. (h) The corresponding size distribution histograms.

X-ray photoelectron spectra (XPS) analysis was performed to gain insight into the interaction of YCl₃ with CsPbI₃ nanocrystals (NCs), and the results are presented in Figure 3(a). The characteristic XPS signals for Cs 3d, Pb 4f, I 3d, Y 3d, and Cl 2p were observed in YCl₃-doped CsPbI₃ NCs and N 1s, O 1s, and C 1s signals coupled with native ligand bonding. The high-resolution XPS spectra of Cs 3d, Pb 4f, I 3d, Y 3d, and Cl 2p are displayed in Figure 3(b-f), respectively. The partial substitution of I⁻ ions by Cl⁻ ions is evidenced by an increase in the binding energies of Pb²⁺ 4f_{5/2} and Pb²⁺ 4f_{7/2} from 143.56 and 138.37 eV to 143.64 and 138.7 eV, respectively. The binding energy of I 3d and Cs 3d display little variation compared to YCl₃:CsPbI₃ nanocrystals (NCs). More importantly, the Y³⁺ and Cl⁻ binding energy signals can be observed in the YCl₃-substituted CsPbI₃ nanorods (NRs). These findings support the partial substitution of I⁻ ions by Cl⁻ ions [25].

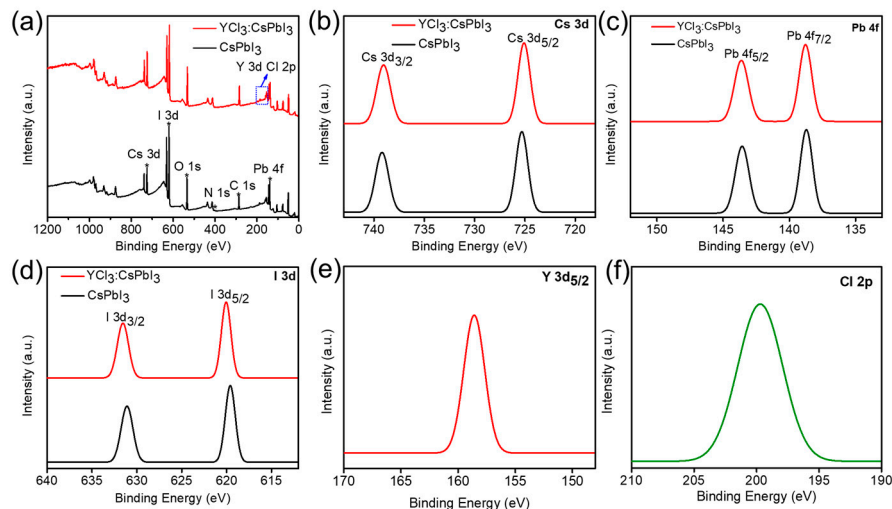


Figure 3. (a) X-ray photoelectron spectroscopy (XPS) of CsPbI₃ NCs and YCl₃:CsPbI₃ NRs. High resolution-XPS spectra of (b) Cs 3d, (c) Pb 4f, and (d) I 3d of pristine CsPbI₃ NCs and YCl₃:CsPbI₃ NRs. (e-f) Y 3d and Cl 2p spectra of YCl₃-substituted CsPbI₃ NRs thin film.

To gain a better understanding of the effects of partial substitution of Pb²⁺ cation and I⁻ with larger radius by the smaller Y³⁺ and Cl⁻ ions, the optical properties of both as-synthesized CsPbI₃ nanocrystals (NCs) and YCl₃:CsPbI₃ nanorods (NRs) were analyzed (Figure 4). The corresponding normalized PL and absorption spectra of YCl₃:CsPbI₃ NRs showed that peak position exhibited a blue-shift owing to partial substitution of Y³⁺ cation [26]. The enlarged bandgap caused the blue-shifts

of the absorption and PL spectra for $\text{YCl}_3\text{:CsPbI}_3$ NRs due to the partial substitution of Pb^{2+} cation and I^- with bigger radius by the smaller Y^{3+} and Cl^- ions [27]. The photoluminescence quantum yield (PLQY) increased from 51% to 70% for the 0.184 mmol YCl_3 passivated CsPbI_3 NCs, suggesting enhanced radiative recombination followed by yttrium chloride doping (**Figure 4c**). The ambient stability of the $\text{YCl}_3\text{:CsPbI}_3$ solution was noticeably improved (**Figure 4d**), the YCl_3 -substituted CsPbI_3 NRs maintained (50% out of 70%) a PLQY, loss of 28% PLQY after being stored for 45 days under ambient condition, prized for the effectiveness of yttrium chloride passivation. However, over the same period of time, the PL quantum yield of pristine CsPbI_3 NCs nearly approached zero. The inset of **Figure 4d** shows the images recorded at different times for the unsubstituted and YCl_3 -substituted CsPbI_3 NCs solution.

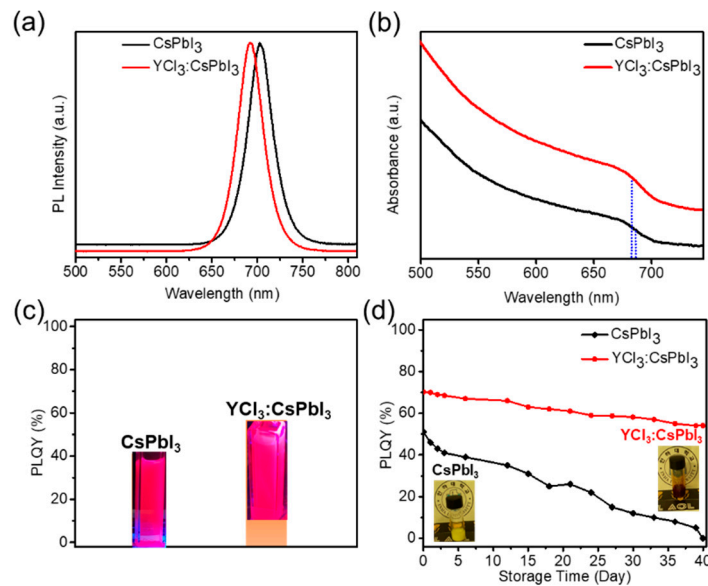


Figure 4. (a-c) Photoluminescence (PL), absorption spectra, and PL-quantum yield (PLQY) of the unsubstituted and YCl_3 -substituted CsPbI_3 NCs. The inset **Figure 3(c)** is the image of the CsPbI_3 NCs and $\text{YCl}_3\text{:CsPbI}_3$ NRs solution under the UV lamp. (d) PLQY values as a function of stored days for unsubstituted and YCl_3 -substituted CsPbI_3 NCs solution. The inset is real images of both samples after 45 days.

The control and YCl_3 -substituted CsPbI_3 NCs were employed as emitters to evaluate the potential applications in perovskite LEDs. The LEDs were fabricated based on the configuration of indium tin oxide (ITO)/ poly(3,4-ethylenedioxythiophene) polystyrenesulfonate (PEDOT: PSS)/poly(4-butylphenyl-diphenyl-amine) (P-TPD)/ unsubstituted CsPbI_3 or $\text{YCl}_3\text{:CsPbI}_3$ / 1,3,5-tris(1-phenyl-1H-benzimidazol-2-yl)benzene (TPBi)/ lithium fluoride (LiF)/ aluminum (Al). The schematic illustration of PeLED, and the corresponding energy levels diagram of the functional layer are illustrated in **Figure 5a-b**. The functional layers' energy level values are taken from previous literature [12]. The thickness of ITO (150 nm), PEDOT: PSS, P-TPD (60 nm), $\text{YCl}_3\text{:CsPbI}_3$ (55 nm), TPBi (60 nm), and LiF/Al (130 nm) were analyzed by the cross-sectional TEM image, as shown in **Figure 5c**. The current density-voltage-luminance (J - V - L) curves of the unsubstituted and YCl_3 -substituted CsPbI_3 NCs LEDs with a 4 mm² emitting area are displayed in **Figure 5d**. The turn-on voltage (where the luminance achieved 1 cd/m²) is reduced from ~3.9 V to ~3.6 V for the YCl_3 -substituted CsPbI_3 NCs LED, revealing that more balanced carrier injected owing to their matched-energy level with carrier transfer layer and enhanced conductivity induced by the YCl_3 -substitution that facilitated the efficient charges injection [12,23]. The CsPbI_3 NCs and $\text{YCl}_3\text{:CsPbI}_3$ NRs-based LEDs showed a maximum luminance of 263.1 cd/m² and 421.8 cd/m², respectively. The electroluminescence (EL) spectra of the unsubstituted and YCl_3 -substituted CsPbI_3 NCs LEDs were observed at 691 nm and 688 nm (**Figure 5e**). The $\text{YCl}_3\text{:CsPbI}_3$ -based LEDs revealed high color purity with Commission

internationale de l'éclairage (CIE) coordinates of (0.71, 0.26), which corresponds to the BT. 2020 color gamut, as shown in **Figure 5f**. The EQE *vs.* luminance curves are displayed in **Figure 5g**. The peak EQE of $\text{YCl}_3\text{:CsPbI}_3$ is 3.16%, 1.86-fold higher than the pristine CsPbI_3 NCs (1.69%) based LED. This enhancement is attributed to enhanced PLQY and more balanced carrier transfer in the $\text{YCl}_3\text{:CsPbI}_3$ EML layer.

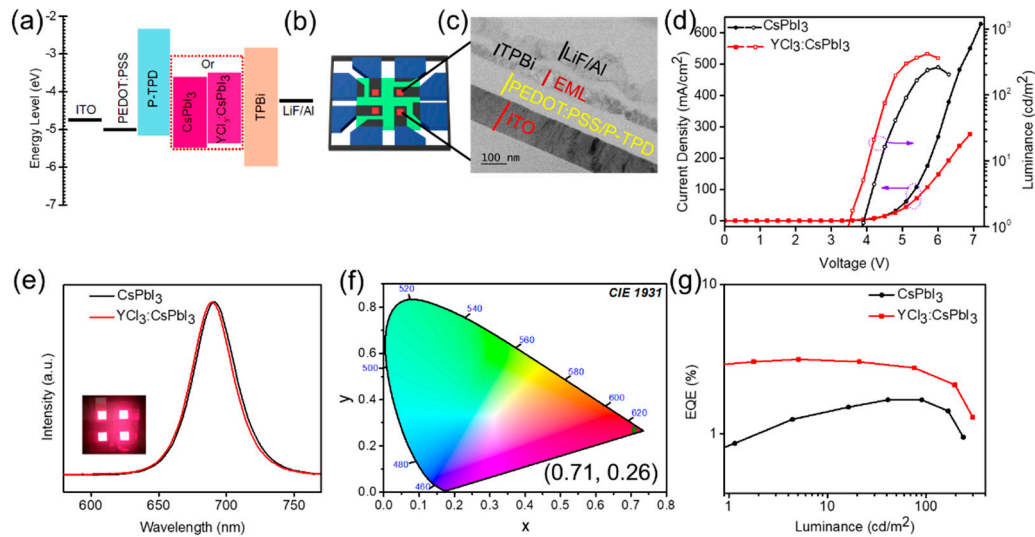


Figure 5. (a) Schematic flat-band energy diagram of perovskite LEDs. (b-c) Schematic device illustration and the corresponding cross-sectional TEM image of the perovskite LEDs. (d) Current density-voltage and luminance-voltage curves, (e) EL spectra of the unsubstituted and YCl_3 -substituted CsPbI_3 LED device. The inset shows the image of a working YCl_3 -substituted CsPbI_3 LED device. (f) The corresponding CIE coordinates for the EL spectrum (0.71, 0.26) of YCl_3 -substituted CsPbI_3 LED. (g) EQE-Luminance curves of the unsubstituted and YCl_3 -substituted CsPbI_3 LED device.

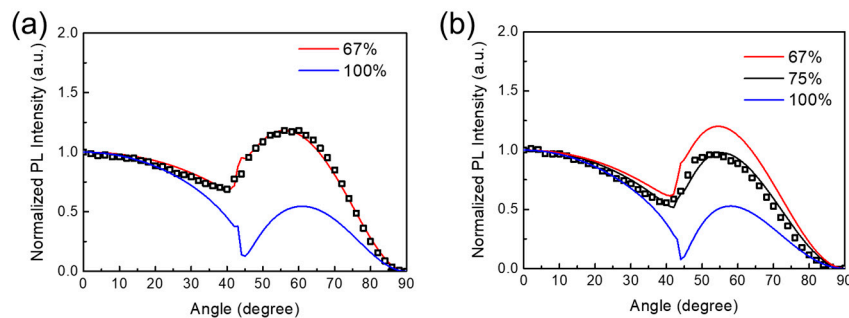


Figure 6. Angle-dependent PL measurements of the perovskite film on a quartz substrate/nanocrystal or nanorods. The experimental data (black squares) are fitted by the classical electromagnetic dipole model, giving a horizontal TDM ratio of (a) CsPbI_3 NCs 67%, and (b) YCl_3 -substituted CsPbI_3 NRs 75%.

To further investigate the enhanced EQE in the YCl_3 -substituted CsPbI_3 NRs, we conducted angle-dependent photoluminescence (ADPL) measurements to probe the orientation of transition dipole moments (TDMs) in the assembled thin film of CsPbI_3 nanocrystals/nanorods. The outcoupling efficiency in PeLEDs has the potential for improvement by controlling the orientation of TDMs [30]. The optical TDMs of nanoplatelets and nanorods are highly anisotropic, and outcoupling efficiency in planar PeLEDs is profoundly associated with the orientation of emissive TDMs [15,17]. The orientation of optical TDMs of the unsubstituted and YCl_3 -substituted CsPbI_3 NCs was measured by the ratio of horizontal TDMs (Θ). The experimental data are fitted to the pattern simulated employing the classical dipole radiation model [31–34]. We confirmed that the Θ values of CsPbI_3

NCs and $\text{YCl}_3\text{:CsPbI}_3$ NRs film are determined to be 67% and 75% (**Figure 6a-b**). The Θ value in anisotropic nanorods is considerably higher than that of isotropic nanocrystals. Thus, the optical TDMs that are horizontally oriented in anisotropic nanorods are preferred for light outcoupling, resulting in the EQE of the LEDs.

4. Conclusions

In this study, we investigated the effects of incorporating metal chloride (YCl_3) into CsPbI_3 nanocrystals (NCs) to control the dimension. We found that incorporating YCl_3 led to a decrease in the lattice parameters of the CsPbI_3 NCs, which resulted from the partial substitution of larger Pb^{2+} and I^- ions with smaller Y^{3+} and Cl^- ions. The presence of Cl^- ions on the surface of the NCs, coupled with the difference in bond energies between Cl^- and I^- ions, led to the anisotropic growth of the CsPbI_3 NCs into one-dimensional (1D) nanorods. The YCl_3 also significantly improved the photoluminescence quantum yield and storage lifetime of the perovskite solution by properly passing nonradiative recombination rates and defects. Finally, we used the YCl_3 -substituted CsPbI_3 nanorods as the emissive layer in red LEDs and observed a significant improvement in their performance. The LEDs exhibited an external quantum efficiency of 3.16% which is 1.86-fold higher than the pristine CsPbI_3 NCs (1.69%) based LED, attributed to the improvement of the ratio of horizontal TDMs in the anisotropic $\text{YCl}_3\text{:CsPbI}_3$ nanorods to 75% from 67% of that of NCs. Overall, the combined characteristics of YCl_3 -substituted CsPbI_3 nanorods show great potential for developing stable and efficient red LEDs.

Author Contributions: M. I. Saleem: Conceptualization, investigation, data analysis, and writing—original draft preparation. A. Katware: Device fabrication. A. Amin: angle dependent PL analysis. S.-H. Jung: *J-V-L* characterization, J.-H. Lee: Conceptualization, writing—review and editing, supervision, funding acquisition. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: Data will be made available upon request.

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Conflicts of Interest: There is no conflict of interest.

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