

High Quantum Yield Colloidal Semiconducting Nanoplatelets and High Color Purity Nanoplatelet QLED

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Abstract—Recently, colloidal semiconductor nanoplatelets (NPLs) have drawn attention due to their quasi-two-dimensional quantum well structure with extremely narrow photoluminescence. However, the quantum yield of NPLs reported so far is still low compared to the traditional semiconductor quantum dots. In this paper, we demonstrate a method of synthesizing CdSe/CdS core/crown NPLs with well-controlled layer thickness [four-monolayer (4-ML)] and aspect ratio, and hence efficiently enhance their photoluminescence quantum yield to $\sim 47\%$ and 72% for the core and core/crown NPLs, respectively. With these NPLs, we further fabricated the 4-ML NPL QLED device emitting at 516 nm with super color purity (linewidth of 10 nm), which is good enough to achieve a wide color gamut larger than Rec. 2020.

Index Terms—Semiconductor nanoplatelets, photoluminescence quantum yield, color gamut, aspect ratio, QLED.

I. INTRODUCTION

THE invention of semiconductor nanocrystals, referred to as colloidal quantum dots (QDs), opens up new field of well-controlled fabrication of low-dimensional quantum material by wet chemistry [1]–[3]. Due to quantum confinement effect, the energy levels inside the QD are uniquely determined by its size. It is in fact a direct experimental verification of quantum

confinement effect. With its unique property not seen in bulk materials, QDs have shown their potential in quantum computation, light-emitting device and quantum detection [4]–[6]. Out of many applications of QDs, using QD as a color-converter in LED lighting or liquid crystal display (LCD) backlight, and electrically driven quantum-dot light emitting diodes (QLED) are the most tangible to commercialization. For display applications, QD is advantageous in emitting in a narrow line spectrum, making a purer color and increasing the color gamut of the display. Meanwhile, the power consumption of the display is reduced significantly. In fact, the so-called QD TV (QD used in LCD backlight) has appeared even in the market. QD has been considered very seriously as the next generation of display technology beyond OLED. Besides size control, shape control provides another dimension to considerably change the electronic structure of quantum nanoparticles of a given material [7]–[11]. For instance, the charge carriers are confined in two dimensions in quantum nanorods, and in one dimension in nanoplatelets (NPLs).

NPLs have been proven to exhibit extremely narrow exciton absorption and photoluminescence spectra [9], [12], [13]. This enables superior color purity of a display device using NPLs as emitters [14]–[16]. Over 30 years of display development, the main stream display has reached 4 k and even 8 k resolution, which has surpassed the limitation of human vision. In other words, there is no need to further increase the resolution of display anymore. However, color has been relatively ignored for a period of time. As we are still using National Television System Committee (NTSC) standard which was introduced in 1953 for CRT. The NTSC standard covers only up to 47% of all the color we can see. With the QD technology, it is possible now for us to significantly increase the color gamut of displays. Wide color gamut (WCG) is becoming the differentiator in display. Ideally, a single QD can emit atomic like narrow emission line, i.e., extremely narrow full-width-at-half-maximum (FWHM) [17], [18]. In reality, size distribution of QDs leads to over-30 nm FWHM. Comparably, due to the well-controlled thickness, NPLs exhibit an ultra-narrow spectrum (FWHM ~ 10 nm), which is not possible for any other known materials or nanocrystals thus far. With such a narrow spectrum, NPLs are promising in bringing about a “color revolution” to display field. With a few years of research, CdSe and CdSe/CdS NPLs synthesis has achieved accurate control over the thick-

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ness, leading to the precise photoluminescence wavelength and narrow FWHM [19]–[22]. Among NPLs of different thicknesses, the 4-ML one shows photoluminescence peaking at ~ 515 nm, which is a relatively good wavelength for green to achieve wide color gamut. However, the 4-ML NPLs are never utilized in the QLED device due to the low photoluminescence quantum yield (PLQY). Reported CdSe and CdSe/CdS NPLs only exhibit less than 20% and 60% PLQY respectively, which are much lower than that of QDs [19], [23]. This greatly limits people's interest in QLED devices based on the 4-ML NPLs.

Here we report the approach to significantly enhance PLQY of CdSe and CdSe/CdS NPLs by precisely controlling the reactant mesh and quantity in order to fabricate high-efficiency QLED device with superior color purity and WCG. In our method, we firstly varied the mesh of Se precursor to tune the lateral aspect ratio of NPLs and achieved a high PLQY of $\sim 50\%$. Followed by this, we further systematically studied the influence of other parameters, including quantities of Se and cadmium acetate precursors on PLQY and lateral size. Based on the fine-tuned high quantum yield CdSe NPLs (as core), we further coated a CdS crown and achieved $\sim 72\%$ PLQY, which is the highest PLQY ever reported for NPLs. We then utilized these 4-ML core/crown NPLs to make a QLED device, which emitted in an extremely narrow spectrum resemble the photoluminescence spectrum with reasonable electroluminescence quantum efficiency. This is the first time ever in reporting a 4-ML NPL QLED device emitting high purity green color.

II. RESULTS AND DISCUSSION

The synthesis of CdSe core NPLs with 4-ML thickness was carried out using a modified recipe [19]. In brief, CdSe core NPLs with well-controlled lateral size and aspect ratio were synthesized as reported but with a different Se mesh (see Experimental for details). Firstly, we started the synthesis of 4-ML NPLs with the generation of CdSe seed particles by using the cadmium myristate and Se precursor from $\sim 130^\circ\text{C}$. The formation of NPLs starts from the addition of cadmium acetate at $\sim 195^\circ\text{C}$, which plays important role in precisely controlling the thickness of NPLs. The vertical thickness and the mean lateral size of the NPLs are characterized by transmission electron microscopy (TEM) (see Fig. 1a, b). The lateral aspect ratio is calculated from the lateral length and width (Fig. S1, Table S1). The synthesized CdSe NPLs have the same vertical thickness (~ 1.2 nm, Fig. S2) corresponding to 4-ML. This is also consistent with the photoluminescence and absorbance peaks in Fig. 1c. We firstly study the significant role of Se in the synthesis of NPLs by affecting the size of CdSe crystal seeds. Different mesh of Se powder is used with the constant amount of cadmium acetate, 70 mg. By using 200 mesh Se as the reactant, we observe NPLs with larger lateral aspect ratio (Fig. 1a). The mean lateral length and width is (27 ± 2) nm and (7 ± 1) nm respectively, and the corresponding aspect ratio is ~ 3.9 . The absorbance spectrum exhibits two peaks: a sharp peak at 511 nm and the broader peak at 482 nm, corresponding to the electron-heavy hole and the electron-light hole transitions, respectively. The photoluminescence spectra show a single narrow peak at 512 nm, resulting from the radiative recombination at

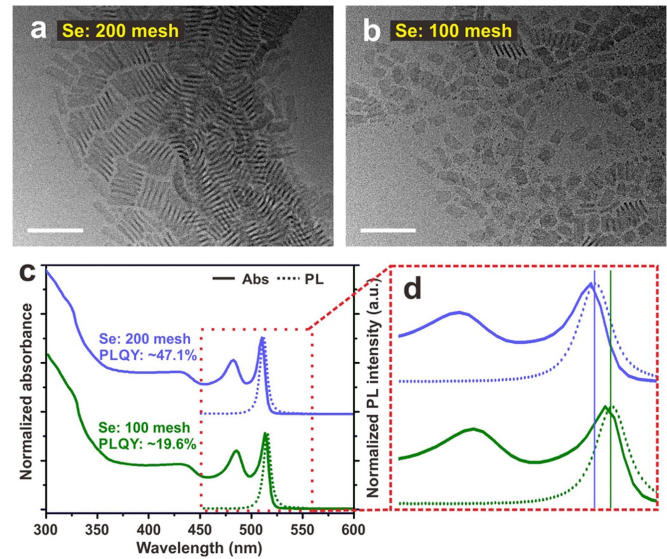


Fig. 1. Synthesis of 4-ML CdSe core NPLs by using Se with different mesh. TEM images of the 4 ML CdSe NPLs by using Se with (a) 200 and (b) 100 mesh. (a) and (b) Scale bar, 50 nm. (c) Absorption and photoluminescence spectra of the corresponding 4-ML CdSe NPLs. (d) Zoom-in spectra from 480–520 nm in (c), showing 3-nm spectral shift of the emission line.

the electron-heavy hole transition, with a very small Stokes shift (~ 1 nm). Significantly, the use of 200 mesh Se achieves the highest photoluminescence quantum yield (PLQY), $\sim 47.1\%$. When Se is reduced from 200 to 100 mesh, the lateral length and width of NPLs also changes to (15 ± 2) nm and (8 ± 1) nm respectively (Fig. 1b). The corresponding aspect ratio is ~ 1.9 . As Se mesh reduces and NPL aspect ratio decreases, for the absorbance spectrum, the sharp peak and the broader peak shifts to 485 nm and 514 nm respectively. The photoluminescence spectrum exhibits a 3-nm shift to 515 nm and PLQY shows a large decay from 47.1% to 19.6%. The full-width-at-half-maxima (FWHM) of the photoluminescence emission peak remains unchanged (~ 8 nm). Detailed information for the absorption and photoluminescence spectra is given in the Table S2.

By tuning Se mesh, we successfully achieve NPLs with high PLQY, which is comparable to that of the traditional quantum dots. This indicates NPLs can realize similar photoluminescent and possibly electroluminescent efficiencies as their QD counterparts. We further studied the influence of precursor quantity on the NPL lateral size and PLQY of NPLs (Table S3). As shown in Fig. 1a, by using a 20 mg Se as the reactant (200 mesh), we observe the generation of NPLs with a lateral aspect ratio (~ 3.9) and high PLQY of $\sim 47.1\%$ (Fig. 2c–e). With the same mesh of Se (200 mesh) and same amount of cadmium acetate (70 mg), an increase of Se amount to 40 mg (200 mesh remained) leads to the change of NPL lateral size. The length and width changes to (29 ± 5) nm and (6 ± 1) nm respectively (Fig. 2a). Hence the aspect ratio exhibits slight enhancement to ~ 5.1 . The corresponding PLQY reduces to 35.7% with the 5-nm blue-shift of photoluminescence peak to 507 nm (Fig. 2c–e). Further increase of Se amount to 60 mg leads to a slight change of NPL lateral size, as well as the generation of NPL with another lateral size. Observed from TEM images, for larger NPLs the lateral length and width is (30 ± 3) nm and (5 ± 1) nm respectively (Fig. 2b),

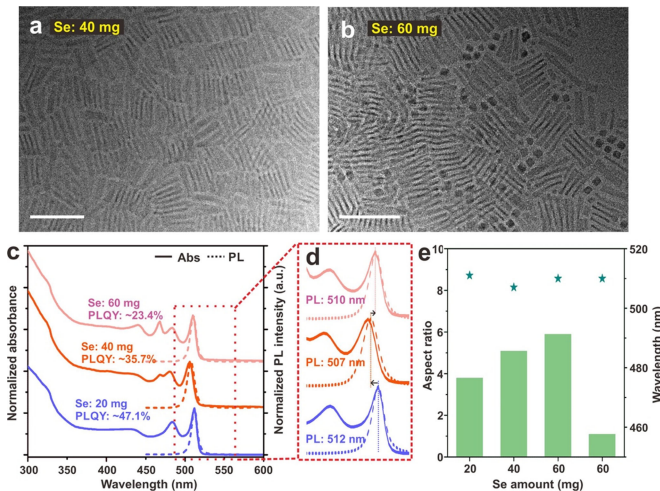


Fig. 2. Synthesis of 4-ML CdSe core NPLs by using 200 mesh Se with different amount. Transmission electron microscopy (TEM) images of the 4 ML CdSe NPLs by using 200 mesh Se with (a) 40 mg and (b) 60 mg. (a) and (b) Scale bar, 50 nm. (c) Absorption and photoluminescence spectra of the corresponding 4-ML CdSe NPLs. (d) Zoom-in spectra from 480–520 nm in (c), showing the spectral shift. (e) The diagram showing the relationship of NPL aspect ratio, photoluminescence peak wavelength and Se amount.

and thus the aspect ratio slightly increases to ~ 5.9 . In contrast, smaller NPLs exhibit much smaller aspect ratio, ~ 1.1 . The single narrow peak in the photoluminescence spectrum proves the generation of homogeneous thickness. However, the two different lateral sizes lead to the splitting of the 2nd peak in the absorbance spectrum. More specifically, the peak at ~ 480 nm was split into two peaks. They are at 468 and 480 nm for 40 mg Se case, and 468 and 484 nm for 60 mg Se case (Fig. 2c). Though we could not see any obvious smaller particles in Fig. 2a as in Fig. 2b, it is clear that such smaller particles were formed as evidenced by its absorbance spectrum. In the 60 mg Se case, the simultaneous increase (~ 5.9) and decrease (~ 1.1) of aspect ratio bring about the 3-nm red-shift of photoluminescence peak back to 510 nm and the corresponding PLQY further decreases to $\sim 23.4\%$ (Fig. 2c–e).

We further studied the effect of cadmium acetate precursor on the synthesis of NPLs. Cadmium acetate plays a significant role in controlling the homogeneity of the thickness of NPLs. Besides, the amount of cadmium acetate also influences the lateral size of NPLs (Table S4). With the constant parameter of Se (200 mesh, 20 mg), different amount of cadmium acetate is used to study its influence. When 40 mg cadmium acetate is added during the reaction, we observe the generation of square NPLs with aspect ratio of ~ 1.1 (Fig. 3a). The corresponding edge length is (8 ± 1) nm. Small amount of cadmium acetate is not able to support the growth of CdSe seed to large NPLs. When cadmium acetate is increased to 60 mg, the lateral size of CdSe NPLs changes to (20 ± 5) nm and (3 ± 1) nm, leading to the strip shape of NPLs. The lateral aspect ratio shows a dramatic increase to ~ 7.1 (Fig. 3b). Further increase of cadmium acetate to 70 mg and 80 mg, the aspect ratio gradually reduces to ~ 3.9 ((27 ± 2) nm and (7 ± 1) nm, Fig. 1a) and ~ 2.8 ((25 ± 3) nm and (9 ± 1) nm, Fig. 3c), respectively. For 90 mg cadmium acetate, the lateral size changes to (21 ± 3) nm and $(12 \pm$

2) nm, with the aspect ratio of ~ 1.8 (Fig. 3d). When we continue increasing cadmium acetate to 110 mg, we observe the co-generation of 4-ML and 5-ML NPLs with similar aspect ratio of ~ 3.4 (Fig. 3e), which is also obvious from the absorbance and photoluminescence spectrum (Fig. S3).

The NPL formation starts by the nucleation of CdSe nanocrystal seeds. During the synthesis, these small nanocrystals gradually disappear with increasingly preferential lateral growth. The NPLs could be formed by self-assembly of the small seeds or by lateral extension of the seed thanks to in situ continuous reaction of Cd and Se precursors (Fig. S4) [12]. Hence the influence of Se mesh is related to the formation of CdSe seeds. Se mesh results in the difference in its reactivity due to the influence on specific surface area and environmental impact. Larger mesh of Se leads to larger specific surface area, which provides higher reactivity for the formation of CdSe seeds. Hence larger mesh of Se results in larger CdSe seeds, leading to a larger lateral size of NPLs. In addition, larger amount of Se leads to the generation of more CdSe seeds. For the same amount of cadmium acetate, more and finer CdSe seeds could lead to the formation of NPLs with smaller lateral size and aspect ratio. However, when the amount of Se exceeds certain value, the same amount of cadmium acetate is not enough to support NPL growth. Part of NPLs are not able to grow to large size and thus two different lateral sizes formed. Furthermore, with the same mesh and amount of Se, as the amount of cadmium acetate increases, more NPL seeds are able to self-assemble or laterally grow, and hence gradually form NPLs with larger size.

Based on the CdSe NPLs with PLQY $\sim 47.1\%$ (as core, Se: 200 mesh, 20 mg; cadmium acetate: 70 mg), we further grew a CdS crown on them and achieved a high PLQY $\sim 71.6\%$, the highest ever reported. The synthesis details are provided in the Experimental section. Fig. 4a exhibits TEM images of core/crown NPL structure with (29 ± 5) nm and (9 ± 2) nm lateral size. The introduction of the CdS crown results in the red-shift of photoluminescence peak to 515 nm (inset image in Fig. 4a). The FWHM of the emission spectra of NPLs still remains very narrow (~ 8 nm) after coating with CdS crowns, which suggests that the NPLs have neither degraded in crystal quality nor into other geometries like spheres or rods, where usually shell-coating results in widened emission. Absorbance of the core/crown structure exhibits the heavy- and light-hole transitions at 515 and 480 nm, respectively. In the case of core-only NPLs, the absorption around 400 nm is flat. However, a new absorption peak at 410 nm starts to arise and becomes more prominent as the CdS crown is grown larger. This peak is due to the first exciton peak of the CdS layer having 4 ML thickness, which also supports the formation of the CdS crown. We further calculated the chromaticity coordinates of the blue 3-ML NPLs (466 nm), proposed green 4-ML NPLs (515 nm), yellow 5-ML NPLs (556 nm), commercial blue InGaN LED (450 nm) [24] and commercially available red quantum dots (630 nm) [25] in Fig. 4b. The positions of 3-, 4-, 5- ML NPLs, blue LED and red quantum dots in CIE 1931 plot are (0.133, 0.044), (0.048, 0.806), (0.348, 0.649), (0.348, 0.649), (0.159, 0.037) and (0.707, 0.293), respectively. The chromaticity coordinates for each point were summarized in the Table S5. Proposed 4-ML

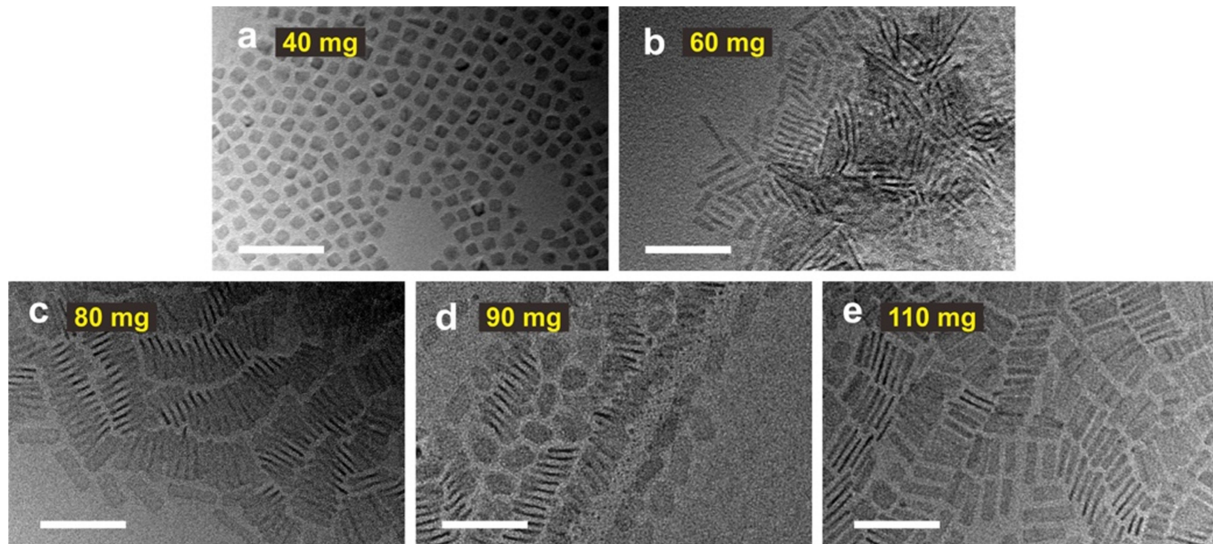


Fig. 3. Synthesis of 4-ML CdSe core NPLs by using cadmium acetate with different amount. Transmission electron microscopy (TEM) images of the 4 ML CdSe NPLs by using cadmium acetate with (a) 40 mg, (b) 60 mg, (c) 80 mg, (d) 90 mg and (e) 110 mg. Scale bar, 50 nm.

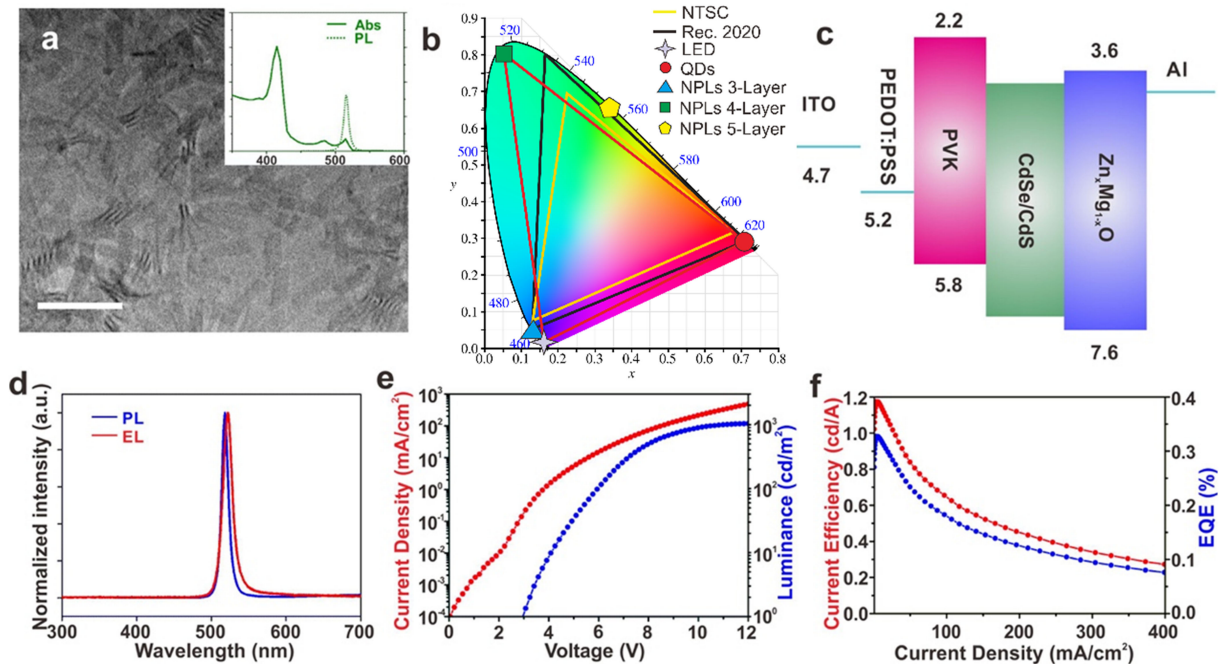


Fig. 4. Fabrication of QLED device by using 4-ML CdSe/CdS core/crown NPLs. (a) TEM images of the 4 ML CdSe/CdS NPLs by using CdSe core NPLs with 47.1% PLQY. Scale bar, 50 nm. Inset is the corresponding absorption and photoluminescence spectra. (b) Chromaticity coordinates for 3-, 4-, 5- ML NPLs and InGaN LED in CIE 1931 color diagram. (c) Energy level diagram for the layered NPL-based QLED devices. (d) Spectral comparison of photoluminescence spectrum of NPL solution versus electroluminescence spectrum of QLED. (e) Current density (J) and luminance (L) versus driving voltage (V). (f) Current efficiency and external quantum efficiency (EQE) versus luminance.

NPLs emitting green light can strongly improve the color gamut of displays, when combined with an InGaN LED [24] for blue and normal QDs [25] for red. The set of color coordinates obtained for our green NPLs combined with color coordinates for blue InGaN LED and for commercial red CdSeS/ZnS alloyed QDs constitute a large triangle in the CIE 1931 color diagram – covering the area, which is 42.2% larger compared to NTSC and 6.2% larger compared to Rec. 2020 color spaces, respectively. With the high quantum yield 4-ML NPLs, we fabricated a

QLED with the device structure as depicted in Fig. 4c, which is consisted of indium tin oxide (ITO)/PEDOT: PSS (30 nm)/PVK (30 nm)/CdSe/CdS core/crown NPLs (40 nm)/Zn_xMg_{1-x}O NPs (40 nm)/Al (100 nm). All layers are spin-coated onto the patterned ITO substrate except for the Al cathode, which is deposited through vacuum thermal evaporation. Fig. 4d shows the PL spectra of CdSe/CdS core/crown NPLs and the EL spectra of the corresponding NPL-based QLED. The green emission of such QLED shows a narrow FWHM (10 nm) with the EL peak

centered at 516 nm. A slightly red-shifted EL peak compared with the PL spectra can be attributed to the combination of Förster energy transfer in close-packed NPL solid films and/or an electric field-induced Stark effect [22]. No noticeable parasitic emission has been observed from the figure, which indicate high purity of the 4-ML NPLs in the sample. Fig. 4e shows the luminance and current density versus applied voltage for the green (at 516 nm) QLED made of CdSe/CdS core/crown NPLs. The device exhibits a turn-on voltage of 2.96 V and a maximum brightness of ~ 1036 cd/m² (at 12 V). The current efficiency and external quantum efficiency (EQE) characteristics of the devices as a function of luminance are plotted in Fig. 4f. The device shows a maximum luminance efficiency of 1.17 cd/A, corresponding to the highest external quantum efficiency of 0.32%. These parameters are still lower compared to their QD counterparts, which is due to, firstly the strong interaction between the neighboring NPLs, resulting in significant energy transfer. Secondly, the plate shape of nanoparticle also makes it difficult to coating a uniform film. However, they are still significantly improved compared to previous reports [13], [22], [26]. This could be a promising approach for the application of QLED devices with super color purity and wide color gamut.

III. CONCLUSION

In this report, we demonstrate a fine-tuned approach to enhance the PLQY of CdSe NPLs by precisely controlling the reactant mesh and amount. In our method, we firstly tune the mesh of Se reactant. By using 200-mesh Se, we successfully achieve the control over the lateral size of NPLs and dramatic enhancement on NPL quality with much higher PLQY, $\sim 47\%$. Followed by this, we systematically study the influence of Se amount and cadmium acetate amount on PLQY and lateral aspect ratio. By increasing Se amount or decreasing cadmium acetate amount, we observe the increment of NPL lateral aspect ratio and reduction of PLQY. Hence by utilizing 20 mg 200-mesh Se and 70 mg cadmium acetate, we obtained the highest PLQY. Based on the high quantum yield of fine-tuned CdSe core NPLs, we further coat CdS crown and achieve $\sim 72\%$ PLQY. Then we further fabricate 4-ML-NPL based QLED device with super color purity and wide color gamut. Due to super-narrow photoluminescence peak of 4-ML core/crown NPLs at 515 nm, we utilize them in the QLED devices with great luminance performance and high electroluminescence quantum efficiency. We believe the NPL technology as demonstrated in this paper provides a promising way for both LCD backlight and for QLED devices, which could bring drastic improvement in color performance to the current displays.

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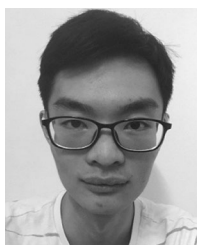
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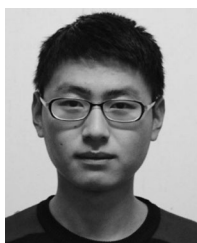
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