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# Eco-friendly quantum-dot light-emitting diode display technologies: prospects and challenges

Peili Gao<sup>1</sup>, Chan Li<sup>2</sup>, Hao Zhou<sup>2</sup>, Songhua He<sup>2</sup>, Zhen Yin<sup>2\*</sup>, Kar Wei Ng<sup>1\*</sup> and Shuangpeng Wang<sup>1\*</sup>

Eco-friendly quantum-dot light-emitting diodes (QLEDs), which employ colloidal quantum dots (QDs) such as InP, and ZnSe, stand out due to their low toxicity, color purity, and high efficiency. Currently, significant advancements have been made in the performance of cadmium-free QLEDs. However, several challenges persist in the industrialization of eco-friendly QLED displays. For instance, (1) the poor performance, characterized by low photoluminescence quantum yield (PLQY), unstable ligand, and charge imbalance, cannot be effectively addressed with a solitary strategy; (2) the degradation mechanism, involving emission quenching, morphological inhomogeneity, and field-enhanced electron delocalization remains unclear; (3) the lack of techniques for color patterning, such as optical lithography and transfer printing. Herein, we undertake a specific review of all technological breakthroughs that endeavor to tackle the above challenges associated with cadmium-free QLED displays. We begin by reviewing the evolution, architecture, and operational characteristics of eco-friendly QLEDs, highlighting the photoelectric properties of QDs, carrier transport layer stability, and device lifetime. Subsequently, we focus our attention not only on the latest insights into device degradation mechanisms, particularly, but also on the remarkable technological progress in color patterning techniques. To conclude, we provide a synthesis of the promising prospects, current challenges, potential solutions, and emerging research trends for QLED displays.

**Keywords:** quantum dots; eco-friendly; light-emitting diodes; degradation mechanisms; displays

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## Introduction

Colloidal quantum dots (QDs) exhibit exceptional properties of narrow-band emission, tunable luminescent wavelength, high luminous efficiency, and remarkable material stability across the visible and infrared spectra, making them highly valuable in diverse applications like imaging, solar collection, displays, communications, and medical<sup>1–6</sup>. The 2023 Nobel Prize in chemistry underscores the immense potential of QDs in basic scientific research, materials science, optics, optoelectronics, and

even biomedical applications. Quantum dot light-emitting diodes (QLEDs) harness the synergies of QDs and organic light-emitting diodes, paving the way for them to emerge as the leading technology in next-generation solid-state lighting and flat panel display<sup>7–9</sup>.

Despite cadmium-based QLEDs having witnessed remarkable progress in recent decades, particularly in red and green QLEDs, challenges persist in improving the stability of blue QLEDs. Additionally, the stringent regulations on heavy metal elements in the European Union

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have posed significant hurdles to the commercialization prospects of cadmium-based QDs. To address this, research has intensified on heavy metal-free QDs, such as Cu-In-Zn-S QDs<sup>10,11</sup>, Ag-In-Ga-S QDs<sup>12–14</sup>, Zn-Cu-Ga-S QDs<sup>15</sup>, and Zn-Ag-Ga-S-Se QDs<sup>16</sup>, which have been synthesized through the incorporation of homologous or heterogeneous ions. However, these QDs still face significant challenges, including a wide full width at half maximum (FWHM) ( $>45$  nm), low external quantum efficiency (EQE) ( $<10\%$ ), and poor device stability ( $<10$  h), significantly hindering their competitiveness in full-color or display applications.

Indium phosphide (InP) QDs, as a substitute for cadmium selenide (CdSe) QDs, have been extensively studied to develop cadmium-free QLEDs<sup>17–21</sup>. The types of eco-friendly QDs are mainly divided into binary (heavy-metal-free group II-VI and III-V) system, multinary (heavy-metal-free group I-(II)-III-VI) system. Currently, the red InP-QLEDs, being eco-friendly, have exhibited outstanding performance, rivaling that of traditional CdSe-QLEDs, in terms of efficiency (EQE=21.4%) and lifetime ( $T_{95}=615$  h@1,000  $\text{cd}\cdot\text{m}^{-2}$ , i.e., the time taken for the brightness to decay to 95% at an initial brightness of 1,000  $\text{cd}\cdot\text{m}^{-2}$ )<sup>22</sup>. Similarly, the green InP-QLEDs exhibit an EQE of 16.3% and a half-lifetime of 5944 hours ( $T_{50}=5944$  h@100  $\text{cd}\cdot\text{m}^{-2}$ , i.e., the time taken for the brightness to decay to 50% at an initial brightness of 100  $\text{cd}\cdot\text{m}^{-2}$ )<sup>23</sup>. However, the utilization of InP for blue QLEDs remains challenging due to its narrow intrinsic bandgap of 1.35 eV, and although blue light emission can be achieved by controlling the particle size within 1–2 nm, this poses immense difficulties in the thermal injection synthesis process. Additionally, the instability and susceptibility to oxidation of InP severely hinder its application potential in high-performance blue QLEDs<sup>24–27</sup>.

Especially, by alloying zinc telluride (ZnTe) with a narrower bandgap of 2.25 eV and zinc selenide (ZnSe) with a wider bandgap of 2.70 eV, the resulting ZnSeTe QDs are considered the most promising candidates for cadmium-free blue QDs<sup>28</sup>, offering flexible adjustment of the bandgap and spectral properties by manipulating the Te/Se ratio. Moreover, the adoption of a quasi-type II band structure can effectively weaken the quantum confinement effect within the ZnSeTe QDs core, suppressing low-energy tail emissions, which contributes to achieving a narrow FWHM, high color purity, and a high photoluminescence quantum yield (PLQY) for blue light emission<sup>28</sup>. Unfortunately, by increasing the Te content in ZnSeTe QDs, their emission wavelength can also be tuned to emit green light (510–530 nm) and even red light (608–612 nm), accompanied by issues such as strain at the QD core-shell interface and broadening of the FWHM<sup>29,30</sup>. Hence, harnessing the merits of InP QDs and ZnSeTe QDs while overcoming their limitations could pave a viable route for the advancement of cadmium-free, full-color QLED devices, unlocking novel prospects for future display technologies.

As seen in Fig. 1, Nozik's team first synthesized InP QDs as early as 1994 by heating a trioctylphosphine oxide precursor solution at 270 °C for several days<sup>31</sup>. In 2000, Banin's research group reported core/shell semiconductor nanocrystals, providing an effective solution to address the non-radiative decay of QDs excited states<sup>32</sup>. It was until 2011 that Lee announced the first green InP-QLED with an EQE of just 0.01% and an FWHM of 70 nm<sup>33</sup>. However, the development of ZnSe-QDs lagged. In 2008, A. Pron's team achieved ZnSe-QDs with tunable photoluminescence within the spectral range of 390–440 nm and an emission FWHM of around 15 nm through the direct reaction of zinc stearate with

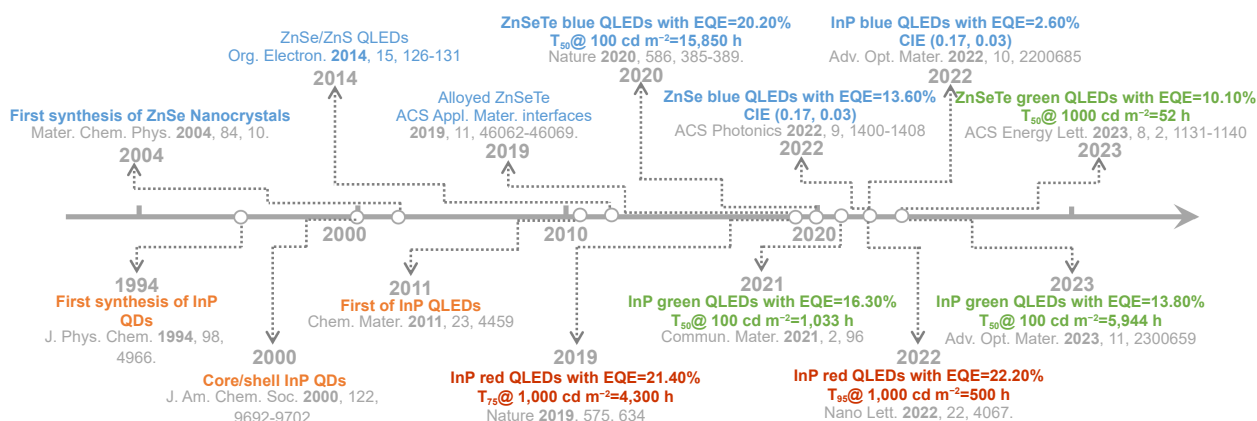


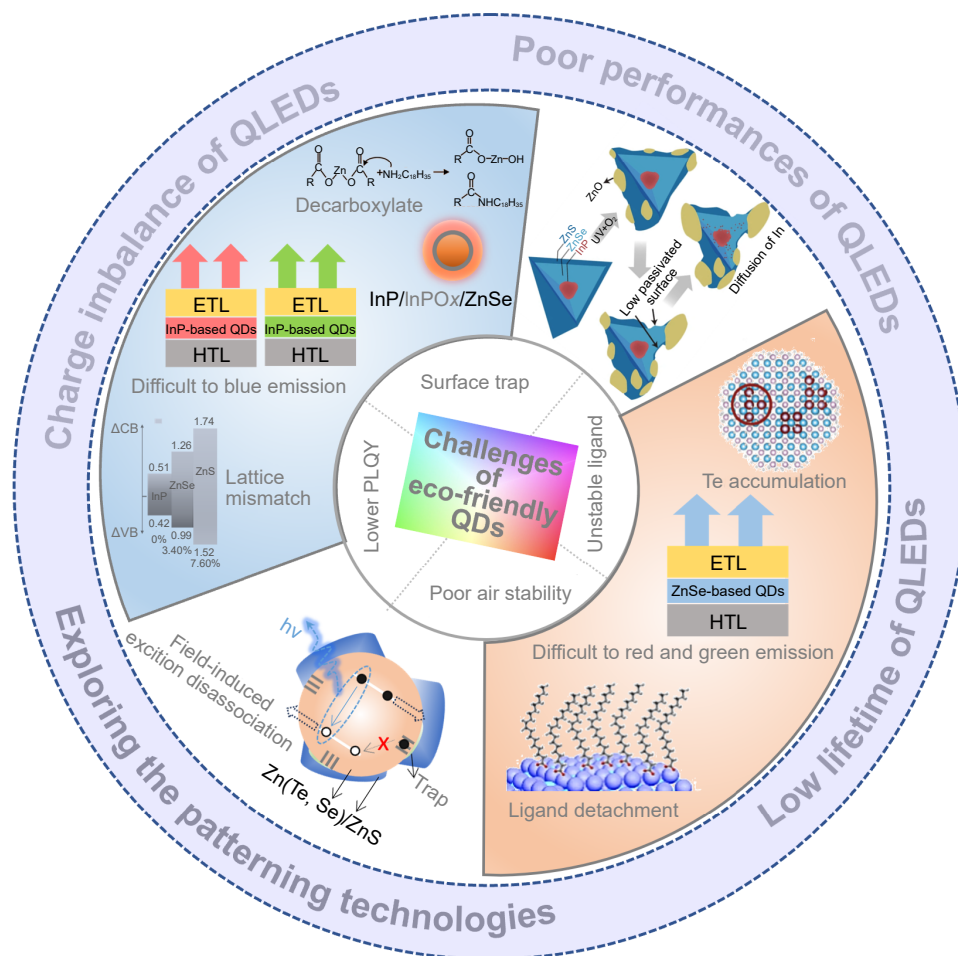
Fig. 1 | Sequential chart of key achievements in the evolution of eco-friendly QLEDs.

selenium dissolved in trioctylphosphine<sup>34</sup>. In 2014, Wedel presented the first blue ZnSe-QLED, exhibiting a maximum brightness of about  $25 \text{ cd}\cdot\text{m}^{-2}$  and an efficiency of approximately  $0.02 \text{ cd}/\text{A}^{35}$ . In 2019, Jang's research group debuted a blue QLED based on alloyed ternary ZnSeTe QDs, demonstrating a peak brightness of  $1195 \text{ cd}\cdot\text{m}^{-2}$  and an EQE of 4.2%<sup>36</sup>. Nevertheless, when the initial brightness reached  $200 \text{ cd}\cdot\text{m}^{-2}$ , the brightness decayed to 50% within only 5 minutes. Fortunately, cadmium-free InP-QLEDs and ZnSe-QLEDs have undergone significant development in the past five years, with cadmium-free red, green, and blue QLEDs achieving EQEs exceeding 15% and half-lifetime up to 1000 hours. Notably, cadmium-free blue ZnSeTe QDs, boasting their superior performance, durability, and color purity, offer promising solutions to the current obstacles hindering the development of blue QLEDs in current enterprises.

Figure 2 clearly illustrates the unique advantages and inherent limitations of InP-based QDs and ZnSe-based QDs. Notably, a shared cause of surface defects in both is

the steric hindrance of long-chain oleic acid ligands and the incomplete passivation of some surface sites during shell formation due to redox reactions, as well as the relatively weak interaction between QDs and ligands. Furthermore, a high PLQY does not inherently guarantee long-lifetime QLEDs. Hence, a thorough analysis of multiple factors, including the photoelectric properties of QDs, the stability of carrier transport layer, and device structure, provides a solid foundation for exploring the similarities and differences among QDs in order to comprehensively enhance QLED performance.

Herein, we review the progress in the field of eco-friendly QD materials and device technologies. Special attention is given to the promising InP-based QDs and ZnSe-based QDs, including their current status, advantages, and inherent limitations. With regard to the challenges in achieving long-lifetime, cadmium-free, full-color QLEDs, we have proposed practical and effective solutions to tackle technical barriers and proposed future research avenues. In particular, this comprehensive review



**Fig. 2** | Key challenges in the development of InP-based QDs and ZnSe-based QDs.



offers an insightful perspective on the intricate relationship between the photoelectric properties of QDs, the carrier transport layer stability, and the device lifetime.

## The working mechanism of QLEDs

QLED is a typical injection-type optoelectronic device that generates electroluminescence under the driving current. The operational process of QLEDs can be simply divided into three stages: (i) the injection and transport of carriers; (ii) the formation and recombination of excitons; and (iii) the emission and extraction of photons. Meanwhile, various factors impact the performance of QLEDs, including carrier balance, interface defects, and device structure.

Especially, the EQE of QLEDs is a crucial parameter to evaluate their performance, and the calculation formula for EQE is as follows:

$$EQE = \gamma \times \eta_{\text{rad}} \times \eta_{\text{out}} \quad (1)$$

The PLQY of the QDs, denoted as  $\eta_{\text{rad}}$ , quantifies the fraction of excitons that undergo radiative decay, which is not only related to the radiative recombination of excitons in QDs but also to the quenching mechanism during the operation of QLEDs. On one hand, the Auger recombination and Förster resonance energy transfer (FRET) within QDs can be effectively suppressed by adopting strategies such as gradient alloyed QDs, thick-shell QDs, and ligand exchange, resulting in a PLQY of up to 100%. On the other hand, the EQE of the devices can be reduced by various processes during operation, including defect-related non-radiative recombination, Auger recombination, and thermally activated defect-assisted recombination.

$\gamma$  represents the charge balance factor, defining the proportion of injected charges that contribute to the formation of excitons. In traditional QLED architectures, the main influential factors of  $\gamma$  are the hole injection barrier, low hole mobility, and electron leakage.

$\eta_{\text{out}}$  denotes the out-coupling efficiency, indicating the percentage of the produced photons that are capable of escaping the device. Since QLEDs feature a sandwich structure composed of stacked multilayers with varying refractive indices, some of the light emitted from the QDs encounters total internal reflection due to the significant refractive index difference between the device layers and the air interface. Typically, only 20%–30% of the total photons can be emitted to the exterior of the device.

Focusing on the key parameters mentioned, various methods have been proposed to significantly enhance the performance of the devices. A comprehensive comparative analysis of these optimized parameters is presented in Table 1, providing a clear insight into the effectiveness of each approach. With improvements in QD core-shell engineering and ligand exchange methods, the PLQY of QDs has reached near-perfect levels of 100%. However, the EQE of QLEDs still remains at around 20%. Meanwhile, it is essential to recognize that a high PLQY does not inherently guarantee a prolonged device life, due to the intricate relationship between the electric properties of QDs, the stability of carrier transport layers, and QLED packaging technology. This article explores the approaches to boost QLED performance in terms of QD architecture, the charge balance of the device, and the intricate aging mechanisms, paving the way for more stable and efficient devices.

## Improving PLQY to enhance the performance of eco-friendly QLEDs

The advancements in QLED research can be categorized into two stages. Before 2015, researchers focused on establishing the QD core-shell structure and creating QDs with superior PLQY. Since 2015, the focus has shifted to optimizing device architectures to enhance efficiency and achieve long lifetimes for industrial applications.

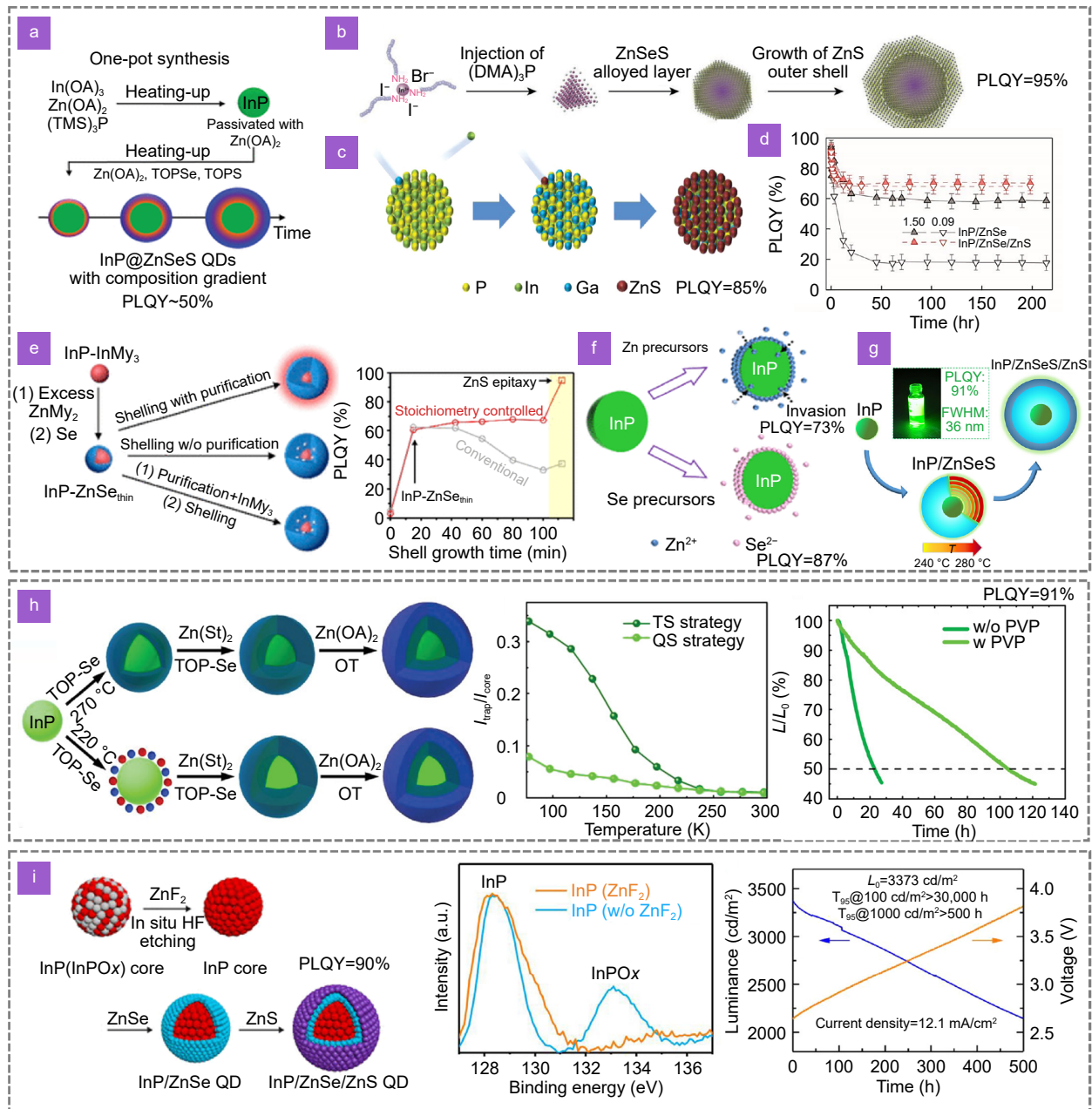
### Core-shell strategies of InP-based QDs

Initially, indium trichloride ( $\text{InCl}_3$ ) and tris-(trimethylsilyl) phosphine ( $(\text{TMS})_3\text{P}$ ) served as the primary phosphorus sources for InP QDs<sup>33</sup>, as depicted in Fig. 3(a). However, the high reactivity of the phosphorus source led to a significant number of defects on the QD surfaces. Additionally, the high reactivity of  $(\text{TMS})_3\text{P}$  not only contributed to an uneven distribution of particle sizes but also resulted in the generation of harmful gases. To address this issue, the H. Yang research team has pioneered the use of  $(\text{DMA})_3\text{P}$  as a substitute for  $(\text{TMS})_3\text{P}$ . The adoption of the more economical and safer  $(\text{DMA})_3\text{P}$  significantly boosted the progress in InP QD research<sup>79,80</sup>. Sun's research discovered that green InP/ZnSeS/ZnS QDs, synthesized with  $(\text{DMA})_3\text{P}$ , exhibit a remarkable PLQY of 95% and an FWHM of 45 nm (Fig. 3(b))<sup>56</sup>.

In 2001, Weller et al.<sup>81</sup> first synthesized core-shell InP/ZnS QDs, which greatly reduced surface defects and consequently boosted band edge emission efficiency.

**Table 1** | Performance of the InP-based QDs and ZnSe-based QLEDs.

| Color | QD architecture           | PLQY/<br>FWHM<br>(nm) | EQE @<br>EL peak<br>(%) | Brightness<br>(cd·m <sup>-2</sup> ) | Lifetime @<br>initial luminance<br>(h)     | Reference                                                        |                    |
|-------|---------------------------|-----------------------|-------------------------|-------------------------------------|--------------------------------------------|------------------------------------------------------------------|--------------------|
| Red   | InP/ZnSe                  | 92/38                 | 22.2 @ 630              | 110,000                             | $T_{95}$ @ 1,000 cd·m <sup>-2</sup> =500   | <i>Nano Lett.</i> 2022, <b>22</b> , 4067                         | ref. <sup>37</sup> |
|       | InP/ZnS                   | 80/43                 | 20.4 @ 628              | 24,080                              | $T_{50}$ @ 100 cd·m <sup>-2</sup> =18      | <i>Nano Res.</i> 2023, <b>16</b> , 7511-7517                     | ref. <sup>38</sup> |
|       | InP/ZnSe/ZnS              | 100/36                | 21.4 @ 630              | 100,000                             | $T_{75}$ @ 1,000 cd·m <sup>-2</sup> =4,300 | <i>Nature</i> 2019, <b>575</b> , 634                             | ref. <sup>22</sup> |
|       | InP/ZnSe/ZnS              | 95/35                 | 18.6 @ 630              | 128,577                             | $T_{75}$ @ 100 cd·m <sup>-2</sup> =107,772 | <i>ACS Energy Lett.</i> 2021, <b>6</b> , 1577-1585               | ref. <sup>39</sup> |
|       | InP/ZnSn                  | 90/-                  | 18.3 @ 613              | 23,451                              |                                            | <i>Nat. Mater.</i> 2022, <b>21</b> , 246-252                     | ref. <sup>40</sup> |
|       | InP/ZnSe/ZnS              | 92/-                  | 18.0 @ 630              | 128,577                             | $T_{50}$ @ 1,000 cd·m <sup>-2</sup> =1,095 | <i>ACS Energy Lett.</i> 2020, <b>5</b> , 3868-3875               | ref. <sup>41</sup> |
|       | InP/ZnSe/ZnS              | 93/42                 | 18.0 @ 630              | 10,000                              |                                            | <i>J. Am. Chem. Soc.</i> 2019, <b>141</b> , 6448-6452            | ref. <sup>42</sup> |
|       | InP/ZnSe/ZnS              |                       | 17.2 @ 630              | 25,000                              | $T_{75}$ @ 2,500 cd·m <sup>-2</sup> =74    | <i>J. Am. Chem. Soc.</i> 2022, <b>144</b> (45), 20923-20930      | ref. <sup>43</sup> |
| Green | InP/ZnSe/ZnS              | /36                   | 16.0 @ 633              | 25,606                              | -                                          | <i>Advanced Electronic Materials</i> 2024, 2400195               | ref. <sup>44</sup> |
|       | InP/ZnSe/ZnS              | 90/34                 | 26.7 @ 543              | 277,000                             | $T_{95}$ @ 1000 cd·m <sup>-2</sup> =1,200  | <i>Nature</i> 2024, <b>635</b> , 854-859                         | ref. <sup>45</sup> |
|       | InP/ZnSe/ZnS              | 91/35                 | 12.7 @ 534              | 175,000                             | $T_{50}$ @ 100 cd·m <sup>-2</sup> =20,000  | <i>Materials Futures</i> 2024, <b>3</b> (2), 025201.             | ref. <sup>46</sup> |
|       | InP/ZnSe/ZnS              | /43                   | 8.5 @ 535               | 18.356                              | $T_{50}$ @ 100 cd·m <sup>-2</sup> =500     | <i>Nanoscale</i> 2023, <b>15</b> , 2837-2842                     | ref. <sup>47</sup> |
|       | InP/ZnSe/ZnS              | 86/39                 | 16.3 @ 545              | 12,646                              | $T_{50}$ @ 100 cd·m <sup>-2</sup> =1,033   | <i>Commun. Mater.</i> 2021, <b>2</b> , 1                         | ref. <sup>23</sup> |
|       | InP/ZnSeS/ZnS             | 97/35                 | 15.2 @ 532              | N/A                                 | N/A                                        | <i>Light Sci. Appl.</i> 2022, <b>11</b> , 162                    | ref. <sup>48</sup> |
|       | InP/ZnSe/ZnS              | 54/43                 | 15.0 @ 538              | 10,010                              | $T_{50}$ @ 100 cd·m <sup>-2</sup> =1,430   | <i>Adv. Opt. Mater.</i> 2022, <b>10</b> , 2202066                | ref. <sup>49</sup> |
|       | InP/ZnSe/ZnS              | 90/36                 | 13.8 @ 539              | 16,788                              | $T_{50}$ @ 100 cd·m <sup>-2</sup> =5,944   | <i>Adv. Opt. Mater.</i> 2023, <b>11</b> , 2300659                | ref. <sup>50</sup> |
|       | InP/ZnSe/ZnS              | 82/34                 | 13.6 @ 531              | 13,900                              | N/A                                        | <i>Chem. Commun.</i> 2019, <b>55</b> , 13299                     | ref. <sup>51</sup> |
|       | InP/ZnSe/ZnS              | 91/36                 | 10.6 @ 540              | 15,606                              | $T_{50}$ @ 100 cd·m <sup>-2</sup> =5,462   | <i>Adv. Sci.</i> 2022, 2200959                                   | ref. <sup>52</sup> |
|       | InP/ZnSe/ZnS              | 87/40                 | 6.2 @ 540               | 6,600                               |                                            | <i>Light: Sci. Appl.</i> 2022, <b>11</b> , 162                   | ref. <sup>48</sup> |
|       | InP/ZnSeS                 | /27                   | 9.6 @ 533               | 40,700                              | $T_{50}$ @ 2,000 cd·m <sup>-2</sup> =1.20  | <i>Adv. Opt. Mater.</i> 2023, <b>11</b> , 2300088                | ref. <sup>53</sup> |
|       | InP/ZnSe/ZnS              | 87/38                 | 9.3 @ 534               | 13,445                              |                                            | <i>ACS Energy Lett.</i> 2022, <b>7</b> , 2247                    | ref. <sup>54</sup> |
|       | InP/ZnSe/ZnS              | 90/49                 | 7.8 @ 535               | 4,955                               | $T_{50}$ @ 100 cd·m <sup>-2</sup> =402     | <i>Adv. Sci.</i> 2022, <b>9</b> , 2200959                        | ref. <sup>52</sup> |
|       | InP/ZnSeS/ZnS             | 95/45                 | 7.0 @ 525               | 1,836                               |                                            | <i>Nano Res.</i> 2021, <b>14</b> , 4243                          | ref. <sup>55</sup> |
|       | InP/ZnSeS/ZnS             | 97/40                 | 6.8 @ 540               | 4,884                               |                                            | <i>Adv. Funct. Mater.</i> 2021, <b>31</b> , 2008453              | ref. <sup>56</sup> |
|       | InP/GaP/ZnS/ZnS           | 30/-                  | 6.3 @ 530               | 3,000                               |                                            | <i>Adv. Opt. Mater.</i> 2019, <b>7</b> , 1801602                 | ref. <sup>57</sup> |
|       | ZnSeTe/ZnSe/<br>ZnSeS/ZnS | 80/45                 | 10.10 @ 523             | 83,556                              | $T_{50}$ @ 1,000 cd·m <sup>-2</sup> =52    | <i>ACS Energy Lett.</i> 2023, <b>8</b> (2), 1131-1140            | ref. <sup>58</sup> |
| Blue  | InP/ZnS/ZnS               | 93/47                 | 2.6 @ 474               | 422                                 |                                            | <i>Adv. Opt. Mater.</i> 2022, <b>10</b> , 2200685                | ref. <sup>59</sup> |
|       | InGaP/ZnSeS/ZnS           | 82/47                 | 2.5 @ 465               | 1,038                               |                                            | <i>Chem. Mater.</i> 2020, <b>32</b> (8), 3537-3544               | ref. <sup>60</sup> |
|       | InP/ZnS/ZnS               | 45/47                 | 1.7 @ 485               | N/A                                 |                                            | <i>Adv. Funct. Mater.</i> 2020, <b>30</b> , 2005303              | ref. <sup>61</sup> |
|       | InP/ZnS                   | 52/49                 | 1.4 @ 483               | 1,162                               |                                            | <i>Adv. Opt. Mater.</i> 2022, <b>10</b> , 2102372                | ref. <sup>62</sup> |
|       | InP/GaP/ZnS               | 81/45                 | 1.0 @ 488               | 3,120                               |                                            | <i>J. Phys. Chem. Lett.</i> 2020, <b>11</b> (3), 960-967         | ref. <sup>63</sup> |
|       | InP/ZnS/ZnS               |                       | 2.1 @ 485               | 165                                 |                                            | <i>Appl. Phys. Lett.</i> 2021, <b>119</b> (22), 221105           | ref. <sup>64</sup> |
|       | ZnTeSe/ZnSe/ZnS           | 100/35                | 20.2 @ 460              | 88,900                              | $T_{50}$ @ 100 cd·m <sup>-2</sup> =15,850  | <i>Nature</i> 2020, <b>586</b> , 385                             | ref. <sup>65</sup> |
|       | ZnSeTe/ZnSe/ZnS           | 90/23                 | 18.6 @ 458              | 4,366                               | $T_{50}$ @ 1,000 cd·m <sup>-2</sup> =0.4   | <i>Chem. Eng. J.</i> 2022, <b>429</b> , 132464                   | ref. <sup>66</sup> |
|       | ZnSeTe/ZnSe/ZnS           | /54                   | 18.2 @ 476              | 22,000                              |                                            | <i>Nano Res.</i> 2023, <b>16</b> , 5517-5524                     | ref. <sup>67</sup> |
|       | ZnSeTe/ZnSe/ZnS           | 95/22                 | 18 @ 452                | 3,520                               | $T_{50}$ @ 100 cd·m <sup>-2</sup> =49.1    | <i>Small</i> 2023, <b>19</b> , 2303247                           | ref. <sup>68</sup> |
|       | ZnSe/ZnS                  | 93/12                 | 13.6 @ 443              | 1,031                               | $T_{50}$ @ 100 cd·m <sup>-2</sup> =305     | <i>ACS Photonics</i> 2022, <b>9</b> , 1400-1408                  | ref. <sup>69</sup> |
|       | ZnSe/ZnS                  | 95/12                 | 12.2 @ 445              |                                     |                                            | <i>Nano Lett.</i> 2021, <b>21</b> , 7252-7260                    | ref. <sup>70</sup> |
|       | ZnSeTe/ZnSe/ZnS           | 100/23                | 10.9 @ 448              | 10,240                              |                                            | <i>Adv. Funct. Mater.</i> 2024, 2313811                          | ref. <sup>71</sup> |
|       | ZnSeTe/ZnSe/<br>ZnSeS/ZnS | 84/27                 | 9.5 @ 445               | 2,904                               |                                            | <i>ACS Energy Lett.</i> 2020, <b>5</b> , 1568-1576               | ref. <sup>72</sup> |
|       | ZnSe/ZnS                  | 69/16                 | 7.8 @ 425               | 2,632                               |                                            | <i>Nanoscale</i> 2015, <b>7</b> , 2951-2959                      | ref. <sup>73</sup> |
|       | ZnSe/ZnS                  | 55/16                 | 6.8 @ 433               | 450                                 |                                            | <i>Small</i> 2020, <b>16</b> , 2002109                           | ref. <sup>74</sup> |
|       | ZnS/ZnTeSe/ZnS            | 85/23                 | 6.8 @ 446               | 14,146                              |                                            | <i>Chem. Mater.</i> 2020, <b>32</b> , 5200-5207                  | ref. <sup>75</sup> |
|       | ZnSeTe/ZnSe/ZnS           | 86/22                 | 5.4 @ 443               | 332                                 | $T_{50}$ @ 120 cd·m <sup>-2</sup> =0.1     | <i>Adv. Mater. Interfaces.</i> 2023, <b>10</b> , 2202241         | ref. <sup>76</sup> |
|       | ZnSe/ZnSe/ZnS             | 70/32                 | 4.2 @ 455               | 1,195                               | $T_{50}$ @ 200 cd·m <sup>-2</sup> =0.1     | <i>ACS Appl. Mater. Interfaces</i> 2019, <b>11</b> , 46062-46069 | ref. <sup>36</sup> |
|       | ZnSeTe/ZnSe/ZnS           | 80/16                 | 4.0 @ 445               | 3,200                               |                                            | <i>Journal Ind. Eng. Chem.</i> 2020, 348-355                     | ref. <sup>77</sup> |
|       | ZnSe/ZnS/ZnS              | 53/14                 | 2.6 @ 444               | 100                                 |                                            | <i>Nanoscale</i> 2021, <b>13</b> , 4562-4568                     | ref. <sup>78</sup> |



**Fig. 3 |** The synthesis strategies for InP QDs. (a) Gradient shell QDs. (b) Multi-shelled QDs synthesized with aminophosphine. (c) Introducing a GaP layer between the InP core and ZnS shell to passivate the surface and eliminate traps. (d) Potential distribution and energy level shifts in core-shell heterostructured QDs with high PLQY. (e) Epitaxial deposition of ZnS on InP QDs. (f) Suppressing the cation exchange at the core/shell interface of InP QDs. (g) Temperature gradient solution growth strategy for the growth of the inner shell layer. (h) A quasi-ZnSe shell to minimize surface defects in the InP core with Ostwald ripening hindrance and lifetime extension. (i) ZnF<sub>2</sub>-assisted synthesis for removing the surface oxide layer of InP QDs to produce high-lifetime QLEDs. Figure reproduced with permission from: (a) ref.<sup>33</sup>, Elsevier; (b) ref.<sup>56</sup>, John Wiley and Sons; (c) ref.<sup>82</sup>, American Chemical Society; (d) ref.<sup>40</sup>, Springer Nature; (e) ref.<sup>42</sup>, (f) ref.<sup>83</sup>, (g) ref.<sup>84</sup>, American Chemical Society. (h) ref.<sup>52</sup>, John Wiley and Sons; (i) ref.<sup>37</sup>, American Chemical Society.

Later, InP/ZnSe and InP/ZnSeS structures were also developed. However, due to the thin shell thickness, a satisfactory solution with a high PLQY and photochemical stability has not yet been achieved. In 2012, Kim et al.<sup>82</sup> introduced a GaP shell that effectively passivated the sur-

face and eliminated traps, while also minimizing the lattice mismatch between the InP core and ZnS shell, as illustrated in Fig. 3(c). Moreover, Bae et al.<sup>40</sup> revealed that the potential distribution and energy level shifts in core-shell heterostructured QDs play a crucial role in their

photophysical properties and charge transport characteristics. By controlling the atomic ratio of QDs to alter the interfacial dipole density, the potential distribution of QDs can be further reshaped. As the interfacial dipole density increases from 0.09 nm to 1.50 nm, the oxidative stability is enhanced, as depicted in Fig. 3(d). In 2019, the Peng group<sup>42</sup> demonstrated that zinc chalcogenides (ZnSe and ZnS) with wide bandgaps can be epitaxially deposited onto InP QDs, effectively confining photo- and electro-generated electron-hole pairs (excitons) primarily within the InP cores. This results in a remarkable PLQY of up to 95%, as shown in Fig. 3(e).

Additionally, the cation exchange during the ZnSe shell formation of InP QDs leads to Zn traps in the lattice, which not only act as lattice defects but also increase charge scattering and hinder charge transport due to local band bending. Fortunately, Yang et al.<sup>83</sup> achieved InP/ZnSe/ZnS QDs with a PLQY of up to 87% by constructing a Se-rich shielding layer on the surface of the InP core (Fig. 3(f)). More recently, Li et al.<sup>84</sup> proposed a one-pot synthesis method that employs a temperature gradient solution growth strategy ranging from 240 °C to 280 °C to grow the inner shell layer, enabling the production of high-brightness, narrow-emission InP/ZnSeS/ZnS multi-shell QDs. This approach enables a high PLQY of 91% and a FWHM of 36 nm, as illustrated in Fig. 3(g). Moreover, the Yang group<sup>52</sup> proposed the quasi-shell-growth strategy, which utilizes a quasi-ZnSe shell to minimize surface defects in the InP core through the passivation of in-terminated vacancies. It further hinders the Ostwald ripening of the InP core at high temperatures, enabling the synthesis of InP/ZnSe/ZnS QDs with a remarkable PLQY of 91% and a narrow emission FWHM of 36 nm. As a result, the green QLED achieved a peak EQE of 10.6% and an impressive half-lifetime exceeding 5,000 hours, as shown in Fig. 3(h).

Particularly, the high reactivity of InP towards water and oxygen poses challenges in precisely controlling the QD core-shell interface. Traditional synthesis methods for InP QDs involve the use of carboxylic acid-based precursor solutions or ligands at temperatures above 188 °C. Unfortunately, these carboxylic acids tend to decarboxylate and generate water at high temperatures, leading to the formation of an oxide layer of InPO<sub>x</sub> that hinders the epitaxial growth of the ZnS shell. Jang et al.<sup>22</sup> employed hydrofluoric acid (HF) to eliminate uncoordinated surface dangling bonds and etch away the InPO<sub>x</sub> oxide layer, thereby passivating defects in the InP core and pro-

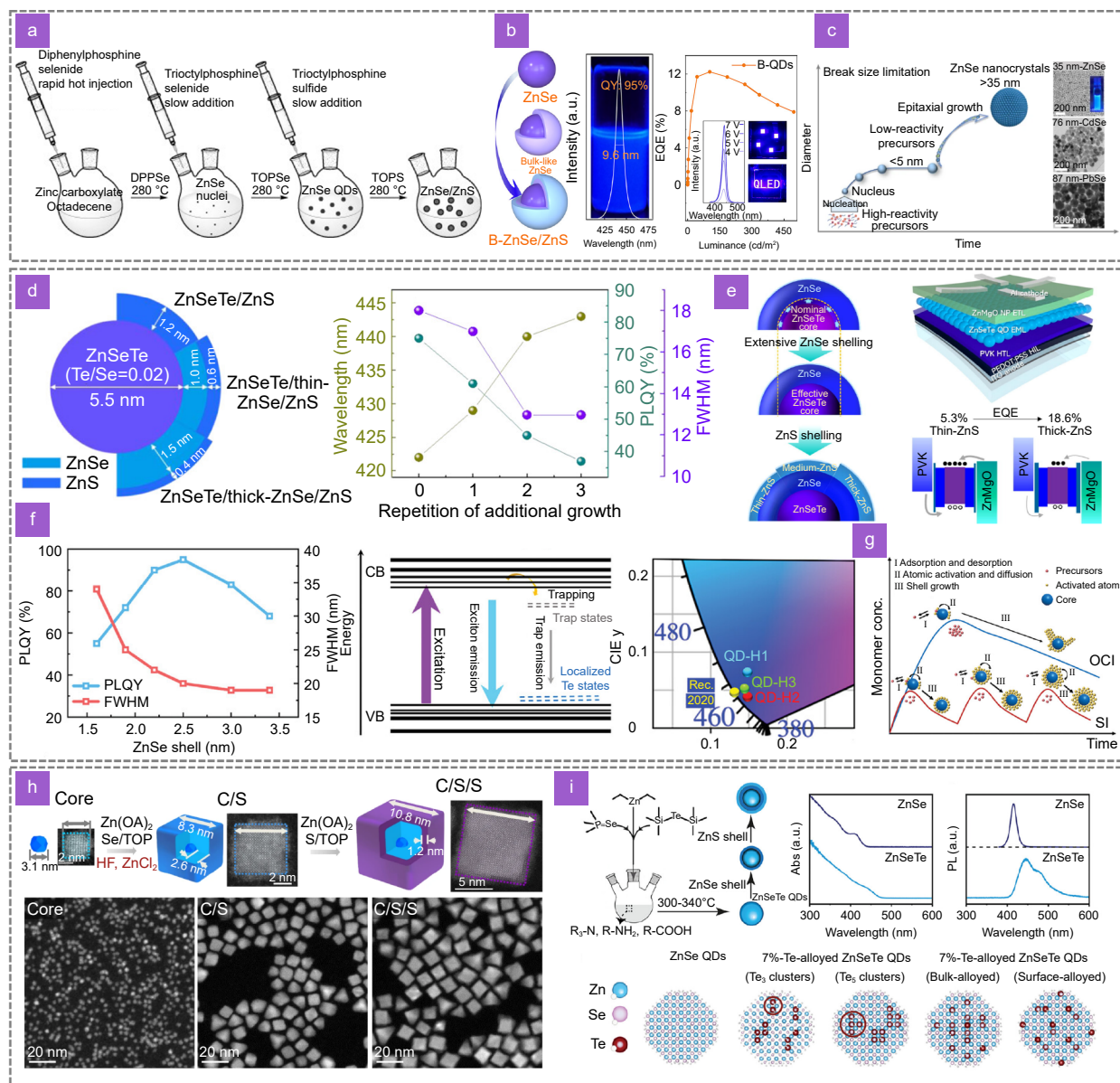
moting a more uniform QD morphology with high PLQY. To reduce the harm caused by HF, Ji's group<sup>37</sup> proposed a method where HF can be generated in situ through the reaction of ZnF<sub>2</sub> with carboxylic acid at high temperatures. This HF then effectively removes the surface oxide layer of InP QDs, enabling subsequent shell growth of ZnSe and ZnS on oxide-free InP QDs. The resulting InP/ZnSe/ZnS QDs exhibit a PLQY of 90% and a FWHM red emission of 36 nm. Furthermore, InP/ZnSe/ZnS QLEDs achieve a remarkable EQE of 22.2% and a T<sub>95</sub> lifetime exceeding 32,000 hours at 100 cd·m<sup>-2</sup>, as depicted in Fig. 3(i). In brief, HF treatment in QD synthesis offers superiorities such as selective etching for purification, controlled size and shape tuning, and enhanced stability. However, it also poses limitations including corrosiveness and safety concerns, environmental impact, complexity in the synthesis process, and limitation in material selection.

### Surface passivation of ZnSe-based QDs

The current reports on eco-friendly QD materials like CuInS<sub>2</sub>, InP, and Si face significant challenges in achieving blue light emission due to the necessity of ultra-small particle diameters, which pose stability issues. In 2014, Wedel's group<sup>35</sup> explored ZnSe QDs for their potential as emissive materials in cadmium-free blue QLEDs. By manipulating the chemical composition during synthesis, they were able to adjust the emission wavelength to fall within the range of 390–435 nm. However, ZnSe QDs primarily exhibit photoluminescence in the violet range, where the wavelengths are insufficient for practical use. Nevertheless, increasing the size of ZnSe QDs can alleviate the effects of quantum confinement, enabling blue light emission, as shown in Fig. 4(a).

Du's group first provide spectroscopic evidence of oxygen contamination on the synthesized ZnSe QDs when they are subjected to oxygen exposure<sup>85</sup>. Meanwhile, they group presented high-quality ZnSe-based core (~5.0 nm)/shell (~1 nm) QDs with a radius greater than the bulk Bohr radius<sup>70</sup>. Through the epitaxial growth of a thin ZnS layer, the ZnSe/ZnS core-shell QDs achieved a PLQY of 95%, and an extremely narrow PL width of around 9.6 nm, as shown in Fig. 4(b). The QLEDs using these bulk-like ZnSe/ZnS core/shell QDs boast a vibrant blue emission centered at 445 nm, a high EQE of up to 12.2%, and a half-lifetime of 237 hours for an initial brightness of 100 cd·m<sup>-2</sup>. Recently, Zhong et al.<sup>86</sup> proposed a novel reaction-controlled epitaxial





**Fig. 4 |** The synthesis strategies for ZnSe QDs. (a) Synthesis scheme for large ZnSe/ZnS QDs. (b) Bulk-like ZnSe core QDs. (c) A reactivity-controlled epitaxial growth strategy. (d) Alloyed ZnSeTe QDs with high PLQY. (e) Heterostructural tailoring of blue ZnSeTe QDs. (f) Controlling the internal ZnSe shell thickness of the QDs. (g) Controlling shell growth of ZnSeTe QDs. (h) Schematic illustrations of the synthesis of ZnTeSe (core), ZnTeSe/ZnSe (C/S) and ZnTeSe/ZnSe/ZnS (C/S/S) QDs, with corresponding TEM images. (i) Influence of Te clustering on optical properties of ZnSeTe QDs. Figure reproduced with permission from: (a) ref.<sup>35</sup>, Elsevier; (b) ref.<sup>70</sup>, American Chemical Society; (c) ref.<sup>66</sup>, Springer Nature; (d) ref.<sup>36</sup>, American Chemical Society; (e) ref.<sup>66</sup>, Elsevier; (f) ref.<sup>68</sup>, John Wiley and Sons; (g) ref.<sup>71</sup>, John Wiley and Sons; (h) ref.<sup>65</sup>, Springer Nature; (i) ref.<sup>68</sup>, John Wiley and Sons.

growth strategy for ultra-large ZnSe nanocrystals, as shown in Fig. 4(c). By injecting precursor monomer solutions with different reactivity at high temperatures and adjusting the injection rate, they successfully suppressed the secondary nucleation during the epitaxial growth process. This was followed by a ZnS shell coating, yielding nearly spherical ZnSe/ZnS nanocrystals with a diameter of 35 nm, which emit blue light with a peak wave-

length of 470 nm and a PLQY of 60%. This strategy provides new insights for synthesizing other types of ultra-large semiconductor nanocrystals. Recently, Wu's research group presented a novel core-shell interface, unintentionally alloyed, which aids in mitigating non-radiative Auger recombination in compact QDs (~7.8 nm)<sup>87</sup>. These QDs show biexciton lifetimes that are comparable to larger CdSe-based systems. Their small size and

prolonged gain lifetime make them incredibly versatile, similar to laser dyes, enabling powerful blue amplified spontaneous emission (ASE) and adjustable laser outputs with a narrow linewidth. This groundbreaking research holds promise for promoting the practical deployment of eco-friendly colloidal QDs in the laser field.

To enhance the standard blue light emission, Yang et al.<sup>36</sup> reported in 2019 the first device based on ZnSeTe QDs (441 nm), achieving a high PL QY of 70% and a FWHM of 32 nm by optimizing the Te/Se ratio and ZnSe inner shell thickness, as displayed in Fig. 4(d). However, it was found that the introduction of Te leads to lattice mismatch issues between ZnSe and ZnTe, and the uneven distribution of Te results in low-energy tail emissions in the spectrum. To address these issues, researchers have focused on controlling interfacial defects and achieving a more uniform distribution of Te. They also precisely tailored the Te/Se ratio of the ZnSeTe core, achieving moderate PL tunability in the deep-blue region<sup>66</sup>. Moreover, a detailed analysis of the ZnS shell thickness variations revealed their influence on QLED operational efficiency, as shown in Fig. 4(e). Devices with thicker ZnS-based QDs exhibited a peak EQE of 18.6%. Nevertheless, their half-life ( $T_{50}@1000\text{ cd}\cdot\text{m}^{-2}$ ) was limited to just 25 minutes. Similarly, Zhao's team<sup>68</sup> refined the ZnSe inner shell thickness to reduce the exciton-LO phonon coupling and trap states in the QDs, as seen in Fig. 4(f). Furthermore, increasing the QD shell thickness suppressed the FRET process in the QD film. Consequently, the ZnSeTe QLED exhibited an EQE of 18% at the 452 nm electroluminescence peak, but the lifetime stands at a comparatively low 49.1 h ( $T_{50}@100\text{ cd}\cdot\text{m}^{-2}$ ). Recently, Tian et al.<sup>71</sup> designed a step-by-step shell growth technique that controls the concentration of monomers and ensures sufficient growth intervals, thereby facilitating the controlled and uniform epitaxial growth of ZnSe and ZnS shells on ZnSeTe cores, as indicated in Fig. 4(g). This approach significantly reduces lattice distortions and defects, thus greatly inhibiting non-radiative charge recombination. The resulting deep blue ZnSeTe/ZnSe/ZnS QDs (448 nm) have a PLQY close to 100%, while the EQE is only 10.9%. Thus, the achievement of both high efficiency and long lifetime in cadmium-free blue QLEDs has proven to be a challenge.

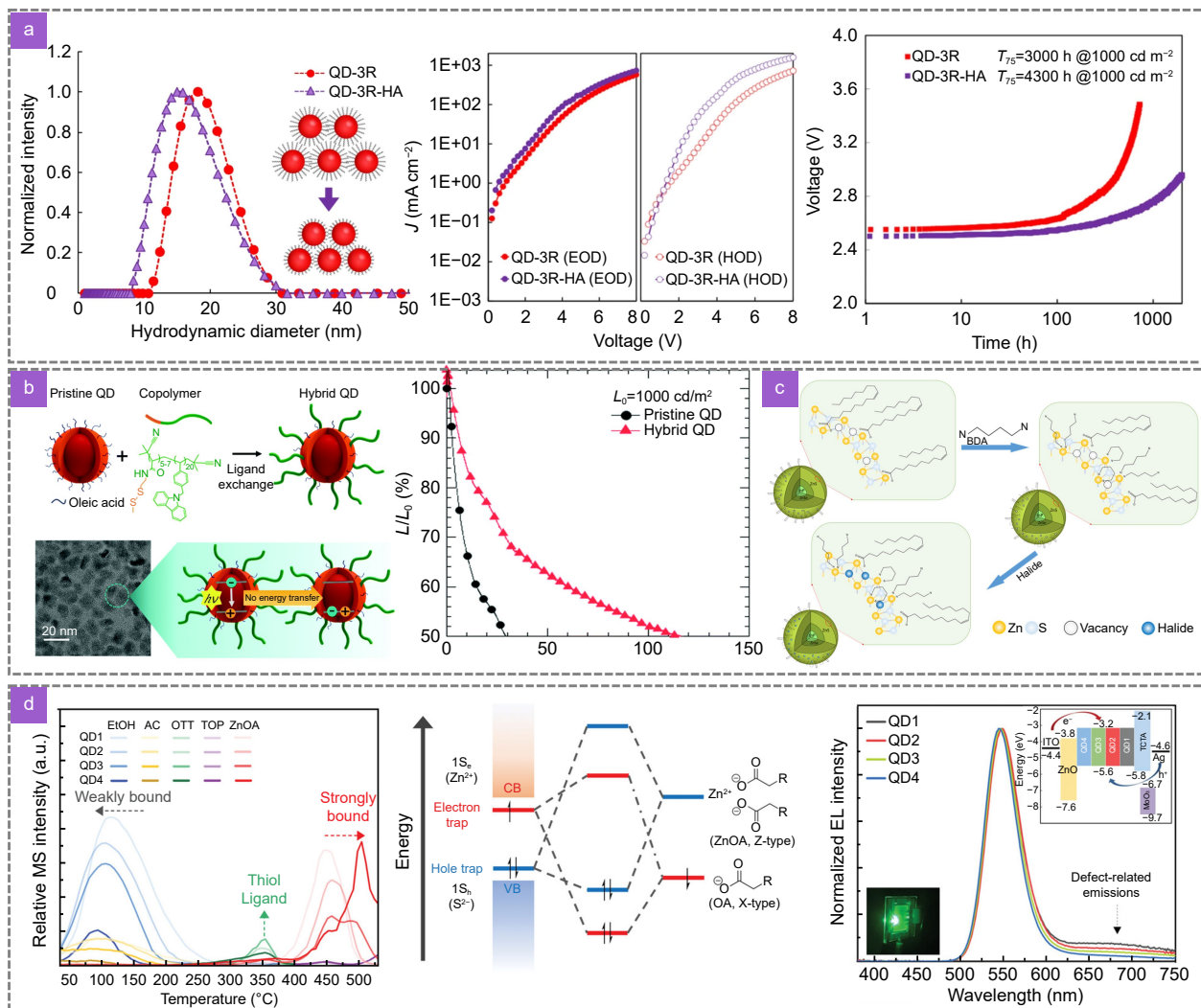
By optimizing the Te doping ratio and the thickness of each shell layer (Fig. 4(h)), Lee's team<sup>65</sup> ultimately achieved tunable emission wavelengths of ZnTeSe/ZnSe/ZnS QDs ranging from 452 to 457 nm,

with PLQY exceeding 90% and FWHM ranging from 23 to 27 nm. It is worth mentioning that the subsequent Cl<sup>-</sup> treatment substitutes the native aliphatic ligands, thereby improving thermal stability and enhancing charge injection/transport. Additionally, the emitting layer features a unique double-stack design with a scaled Cl<sup>-</sup> concentration, which promotes optimal charge recombination. As a result, the device achieves an EQE of up to 20.2% and a  $T_{50}$  of 15850 hours at  $100\text{ cd}\cdot\text{m}^{-2}$ , representing the highest values ever reported for blue QLEDs. Furthermore, to alleviate the hazards of using HF, a novel synthesis approach has been introduced that employs benzoyl fluoride as a chemical additive for the in-situ production of anhydrous HF, as shown in Fig. 4(i). This approach ensures a controlled release of HF during the nucleation and growth stages, leading to a uniform distribution of Te in the ZnSeTe lattices<sup>88</sup>. The resulting core/shell/shell QDs exhibit emission in the natural blue region (457–463 nm), with an FWHM ranging from 22 to 25 nm and a PLQY of  $85\% \pm 5\%$ .

#### Ligand exchange strategy of InP and ZnSe-based QDs

The conventional synthesis of QDs (including InP and ZnSe types) is typically based on the use of carboxylic acid precursor solutions or ligands at high temperatures ( $>188\text{ }^{\circ}\text{C}$ )<sup>13,89–91</sup>. Unfortunately, carboxylic acids undergo decarboxylation at high temperatures, which can lead to the regeneration of defects at the QD core-shell interface, thereby increasing non-radiative recombination channels in QDs<sup>92–94</sup>. Additionally, the feeble interaction between long-chain carboxylic acid ligands and metal ions undermines the chemical stability of QDs<sup>57,95,96</sup>. Moreover, the intrinsic low conductivity of the alkyl chains in carboxylic acids hinders the charge transport properties of QDs<sup>97,98</sup>. To address these issues, researchers have proposed a ligand exchange strategy for QDs, which not only enhances the PLQY of QDs but also achieves long-lifetime QLEDs.

Many ligand exchange preparation strategies exist for InP-based QDs, facilitating the control of their surface properties, stability, and optoelectronic performance, as shown in Fig. 5. For instance, Jang et al.<sup>22</sup> achieved a 63% substitution of long-chain oleic acid with short-chain hexanoic acid (HA), forming QD-3R-HA, which improved the charge injection in QLED. The hole current of short-chain ligands increased by four times, which facilitated exciton recombination and reduced Auger



**Fig. 5 |** The ligand exchange strategies for InP QDs. (a) Replacing oleic acid ligands with hexanoic acid. (b) Substituting oleic acid ligands with semiconducting diblock copolymer units. (c) Modifying InP QDs with different alkyl diamines. (d) Investigating the ligand effect in the passivation of InP QDs. Figure reproduced with permission from: (a) ref.<sup>22</sup>, Springer Nature; (b) ref.<sup>99</sup>, Royal Society of Chemistry; (c) ref.<sup>23</sup>, Springer Nature; (d) ref.<sup>95</sup>, John Wiley and Sons.

recombination, thus inhibiting the voltage increase at the interface between TFB and QDs. As depicted in Fig. 5(a), the fabricated HA-QLED exhibited a more stable operating voltage during device operation. In addition, by dispersing the pristine QDs in chlorobenzene with the aid of a semiconducting diblock copolymer, Kwak's research<sup>99</sup> observed that the copolymer units surrounding the QDs widened the inter-dot distances, minimizing FRET and consequently reducing PLQY. Meanwhile, the carbazole groups within the copolymer contributed to efficient hole transport into the QDs while blocking excessive electron leakage. As a result, the hybrid QLED device exhibited a half-lifetime of 120 hours at an initial brightness of 1000 cd·m<sup>-2</sup>, which is four times longer than that of the unmodified QLED, as seen in Fig. 5(b).

Similarly, the Chou group<sup>23</sup> integrated the InP QDs into a prepared stock solution, which consisted of a mixture of EDA, BDA, or HAD toluene solutions and an anhydrous ethanol solution containing ZnCl<sub>2</sub>, ZnBr<sub>2</sub>, and ZnI<sub>2</sub>. As illustrated in Fig. 5(c), the emitting layer was subsequently treated with passivating agents, which comprised a variety of alkyl diamines and zinc halides. This strategy led to a reduction in electron mobility and an enhancement in hole transport. Additionally, Wedel et al.<sup>95</sup> explored the interplay between the diverse ligands, including zinc octoate, 1-octanoate, zinc stearate, 1-tri-octylphosphine, acetone, ethanol, and 1-octanethiol and the metal ions in InP/ZnSe/ZnS QDs, as illustrated in Fig. 5(d). The findings revealed the presence of solvent ligands, such as ethanol and acetone, on the imperfect

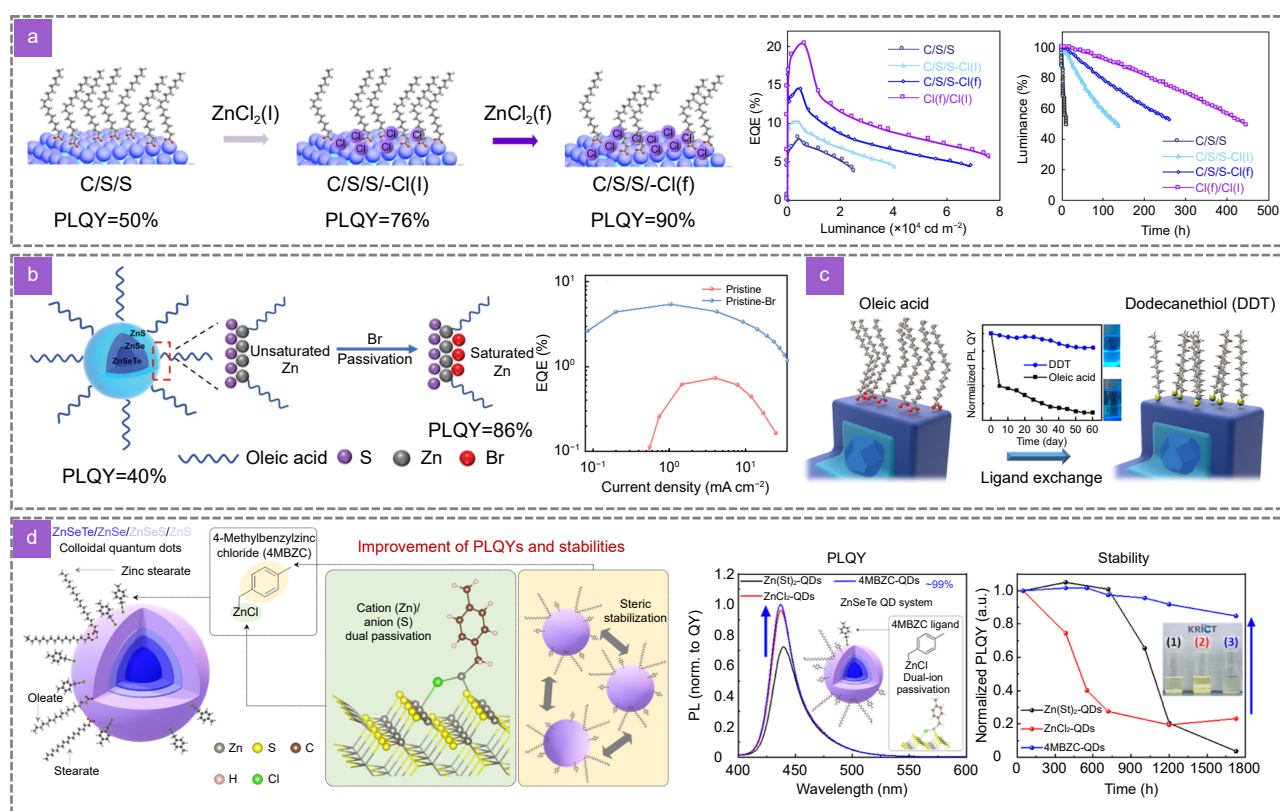


surfaces generated during QDs synthesis, and the carboxylate ligands were mostly present as carboxylates on the QD surface. Furthermore, surface defects in these QDs give rise to electronic states within the bandgap, ultimately hindering the charge transport properties of QLEDs.

Apart from the ligand exchange methods utilized for InP QDs, there exists an extensive suite of preparation protocols specifically tailored for ZnSe QDs, offering new avenues for property improvement. For example, Jang et al.<sup>65</sup> uncovered a mechanism where halogen anions stabilize Zn dangling bonds on the QDs surface upon exchanging ligands with  $\text{ZnCl}_2$ , enhancing their stability. In essence, the PLQY of QDs improved from an initial 50% to 76% through liquid-phase ligand exchange and further to 90% via solid-phase ligand exchange. Meanwhile, the lifetime of QLEDs incorporating ligand-exchanged QDs was extended by nearly ten times, as shown in Fig. 6(a). The bromide decoration technique, as demonstrated by Chen's group<sup>76</sup>, simultaneously tackles QD surface imperfections by the passivation of unsaturated Zn sites, which can enhance carrier radiative re-

combination and eliminate excess oleic acid through ligand exchange, facilitating efficient carrier transport. These benefits lead to a substantial improvement in PLQY, increasing from 39.7% to 86.2%, as shown in Fig. 6(b). Moreover, through the substitution of oleic acid with various alkanethiol ligands, Woo et al.<sup>100</sup> successfully improved the water and oxygen stability, particularly for QDs modified with dodecanethiol (DDT), as illustrated in Fig. 6(c). Upon exposure to air for 2 months, the PLQY of these QDs maintained a high level, decreasing from 94% to 75%.

Despite the excellent surface defect passivation properties of zinc carboxylate and zinc chloride for QDs, they are still plagued by issues such as carboxylic acid ligand detachment and QDs aggregation. To overcome these issues, 4-methylbenzylzinc chloride ligands were utilized on the surface of ZnSeTe/ZnSe/ZnSeS/ZnS QDs, resulting in enhanced PLQY and PL stability<sup>101</sup>. The zinc-chlorine component of the 4MBZC ligand provides dual passivation and strong binding affinities to the QDs surfaces, while the aromatic group promotes stable dispersion in solution. Importantly, the 4MBZC-coated QD



**Fig. 6 | ZnSe QDs ligand exchange strategies.** (a) Chloride passivation via liquid or solid ligand exchange with oleic acid. (b) Bromide decoration of ZnSeTe/ZnSe/ZnS QDs. (c) Oleic acid surface ligand exchange with an alkanethiol variant. (d) Introduction of 4MBZC as a dual-ion passivation ligand. Figure reproduced with permission from: (a) ref.<sup>65</sup>, Springer Nature; (b) ref.<sup>76</sup>, John Wiley and Sons; (c) ref.<sup>100</sup>, (d) ref.<sup>94</sup>, American Chemical Society.



solutions demonstrated remarkable stability, maintaining 84.8% of their initial PLQY (99%) after 1,728 hours of storage, as shown in Fig. 6(d).

## Improving charge balance to enhance the performance of QLEDs

With a profound understanding of QLEDs' operational mechanism, the device structure has undergone significant evolution. Back in 1994, the Alivisatos' group<sup>102</sup> at UC Berkeley pioneered electroluminescence devices incorporating nanocrystals and polymers, initiating a new era in QD lighting, which was categorized as Type-I QLEDs, as exemplified in Fig. 7. With the rapid advancements in QLED technology, three distinct structural variations emerged. Among them, placing the QD emission layer between two organic transport layers gave rise to Type-II QLEDs<sup>103</sup>. Meanwhile, the incorporation of inorganic materials for charge transport led to the designation of Type-III QLEDs<sup>104</sup>. Additionally, the innovative combination of organic and inorganic components was named Type-IV QLEDs, reflecting the synergy between these disparate materials<sup>105</sup>.

In traditional sandwich structures, it is found that excessive electron injection in QLEDs triggers Auger recombination within the QDs. This process not only undermines the stability of the QDs but also causes efficiency roll-off in QLEDs under high current operation. Moreover, the excessive electrons can leak into the HTL, accelerating its aging and thereby impacting the device's lifetime. Meanwhile, the disparity in injection efficiencies between electrons and holes in conventional QLED designs underscores the importance of carrier dynamics optimization. To tackle this, key strategies involve enhancing the injection and transport capabilities of the HTL, optimizing the hole injection efficiency of QDs, suppressing electron injection, and achieving a balanced charge distribution within the device.

## High performance of InP-based QLEDs

### Suppress electron injection and transport

Since 2011, the introduction of ZnO NPs as an ETL in

QLED by Qian et al.<sup>105</sup> has sparked widespread interest in type-IV QLEDs integrating organic and inorganic materials. ZnO NPs possess versatile surface functional groups and allow for straightforward solution synthesis, facilitating diverse chemical modifications. Additionally, various strategies, such as controlling the size of ZnO nanocrystals<sup>106</sup>, metal ion doping<sup>107–110</sup>, and organic functionalization (depicted in Fig. 8(a))<sup>54</sup>, have proven effective in mitigating exciton quenching at QD/ETL interfaces, minimizing ZnO oxygen vacancies, and regulating electron injection, ultimately optimizing charge balance for superior QLED performance. Additionally, dipole moments have proven effective in mitigating defects in ZnO. Yang et al.<sup>111</sup> utilized a dielectric layer composed of phenylethylammonium bromide (PEABr) and methylammonium bromide (MABr), which not only shifts the conduction band minimum of ZnMgO upwards to prevent the injection of extra electrons but also passivates its defect states via Br filling, enhancing charge balance in QLEDs (Fig. 8(b)). Consequently, the QLEDs achieved an operational lifetime of more than 400 hours.

Although ZnMgO thin films are commonly utilized as ETLs in QLEDs for their electron transport properties, they often lead to challenges including interface exciton quenching and excessive electron injection, affecting device performance. In particular, in cadmium-free green QLEDs, our group has found that PO-T2T with its shallower conduction band and lower defect density, outperforms ZnMgO as an ETL, as illustrated in Fig. 8(c)<sup>49</sup>. This not only diminishes interfacial exciton quenching but also improves charge balance. In contrast to traditional organic/QD/inorganic structures, organic/QD/organic architectures demonstrate potential for efficient and stable cadmium-free QLEDs.

### Improve hole injection and transport

To date, considerable efforts have been devoted to tackling the challenge of charge imbalance within QLEDs. For instance, Kwakthe et al.<sup>112</sup> employed CzSi as a hole-blocking interlayer, effectively diminishing the exciton-hole quenching process. Additionally, an ultrathin LiF

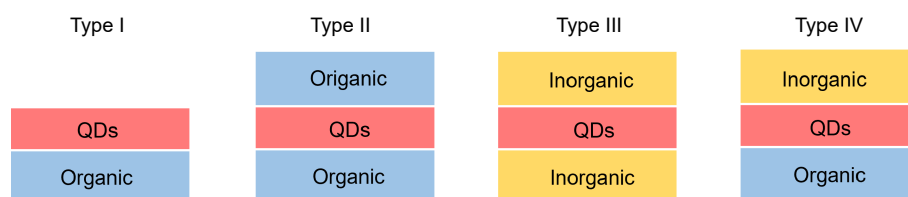
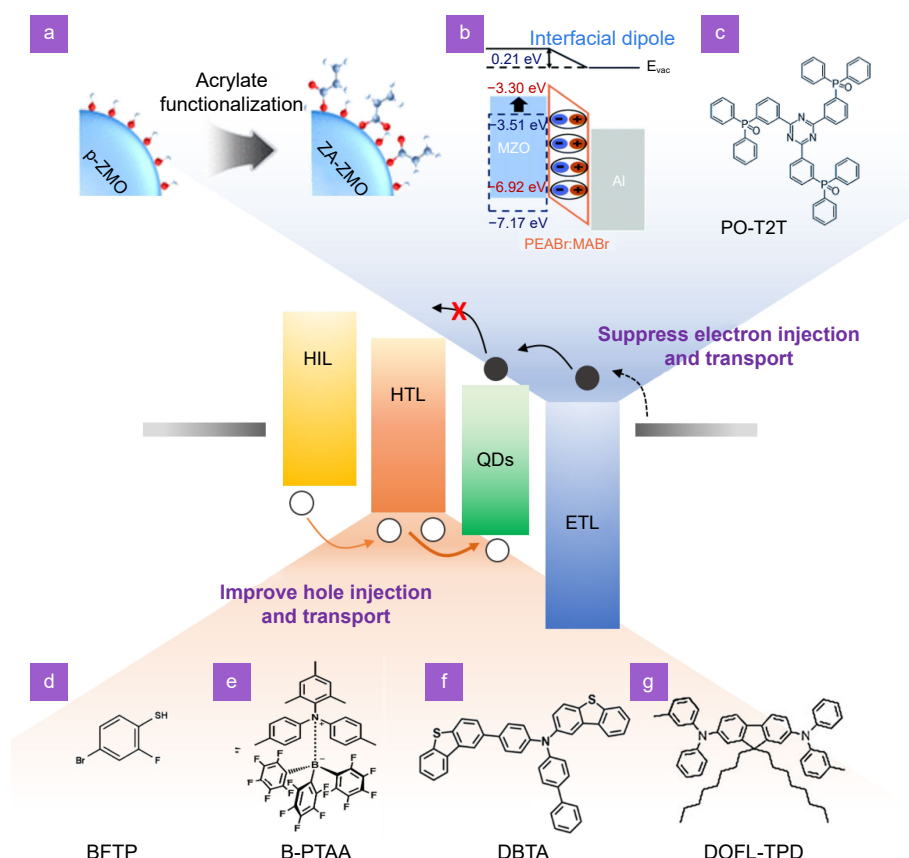


Fig. 7 | Development of QLED device structure.



**Fig. 8 |** The improvement of the charge balance in InP-QLEDs. (a) Acrylate-functionalized ZnMgO. (b) The PEABr:MABr interlayer between ZnMgO and Al cathode. (c) Organic PO-T2T as an ETL to prevent the over-injection of electrons, leading to improved charge balance within the QLEDs. The molecular structure of (d) BFTP-regulated TFB self-assembled dipole interface monolayer, enhancing hole injection efficiency into InP QDs. (e) B-PTAA as an alternative material for HTL with high hole mobility and a deep HOMO level. (f) DBTA utilized as HTL. (g) A blended EML by combining DOFL-TPD with InP-based QD. Figure reproduced with permission from: (a) ref.<sup>54</sup>, American Chemical Society; (b) ref.<sup>111</sup>, Royal Society of Chemistry; (c) ref.<sup>49</sup>, John Wiley and Sons; (d) ref.<sup>47</sup>, Royal Society of Chemistry; (e) ref.<sup>38</sup>, Tsinghua University Press; (f) ref.<sup>41</sup>, American Chemical Society; (g) ref.<sup>39</sup>, American Chemical Society.

layer is employed to tailor the work function of ITO, effectively preventing electron leakage while promoting hole injection<sup>113</sup>. Moreover, Yang et al.<sup>47</sup> developed an energy-level regulated QLED using BFTP self-assembled monolayers on the surface of the TFB, which has a backbone benzene ring and functional head group, as illustrated in Fig. 8(d). The molecular dipole layer at the interface of QDs and HTL accomplishes two key functions: it lessens the energy barrier for hole injection into QDs via vacuum level offset, and it stops the HTL from quenching the fluorescence of QDs. To hinder electron transfer, they recently have also inserted an ultra-thin interlayer of polyvinylpyrrolidone (PVP) between TFB and QD. With the PVP interlayer<sup>52</sup>, parasitic emissions from TFB were eliminated, resulting in QLEDs with a lifespan exceeding five times that of the reference device.

In particular, novel hole transport materials, such as

B-PTAA<sup>38</sup> and DBTA<sup>41</sup> have been developed, which feature rigid dibenzothiophene and tertiary amine units, as illustrated in Fig. 8(e) and Fig. 8(f). These materials offer high hole mobility and a deep highest occupied molecular orbital level, enhancing hole injection efficiency into InP QDs. Furthermore, a homogeneous emitting layer has been created by combining an organic hole-transporting material (DOFL-TPD) with InP/ZnSe/ZnS QDs, as presented in Fig. 8(g). This combination offers exceptional hole mobility ( $1.1 \times 10^{-3} \text{ cm}^2/\text{V}\cdot\text{s}$ ) and optimal LUMO/HOMO energy levels (2.4 eV/5.4 eV), respectively, effectively blocking electron leakage and enabling efficient hole injection into the QD EML<sup>39</sup>.

#### Standard blue light emission of ZnSe-based QLEDs

The disparity in energy band offsets, where the offset between the anode Fermi level and the valence band

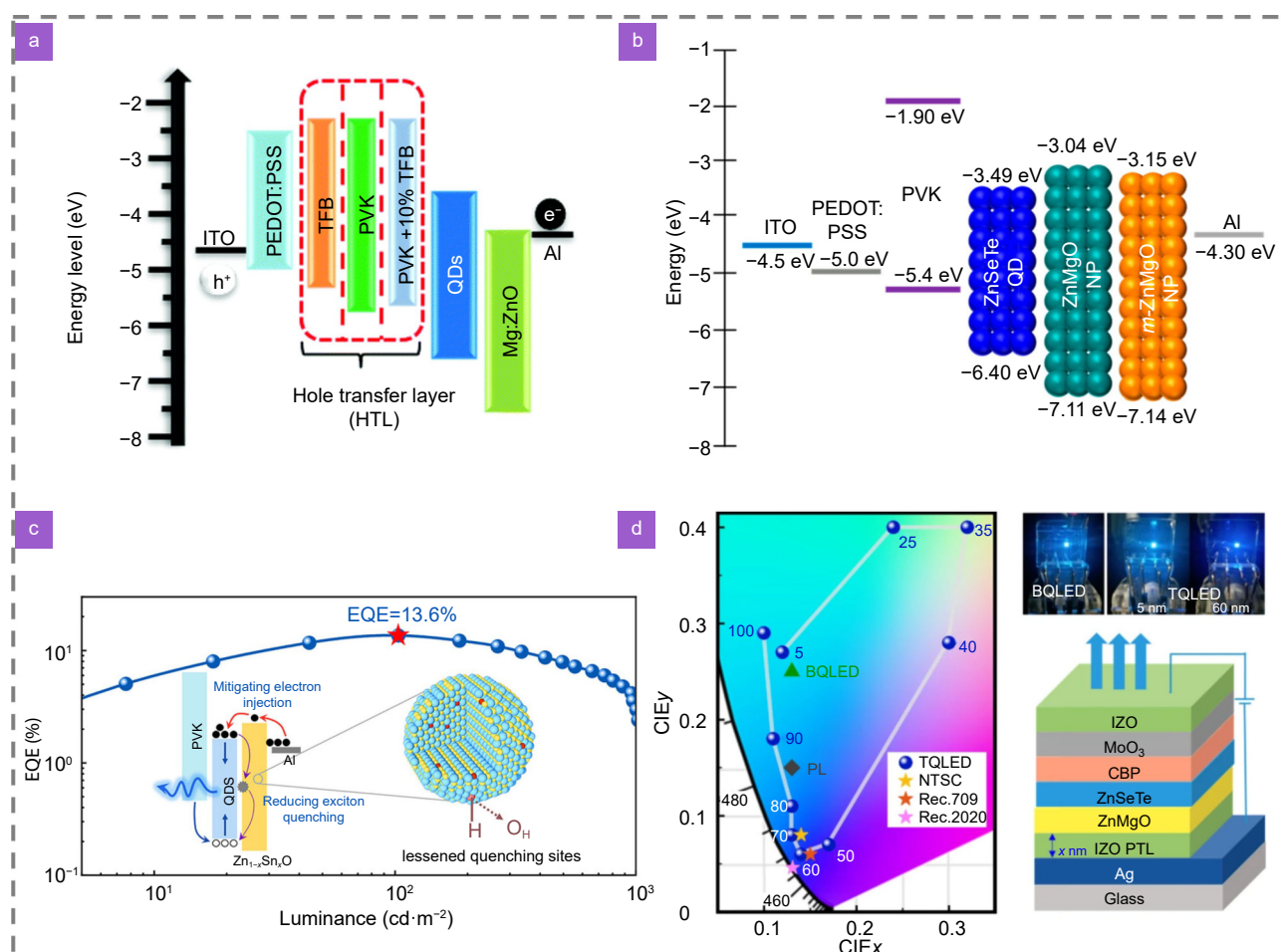
maximum of QDs significantly exceeds that between the cathode Fermi level and the conduction band of QDs, is particularly pronounced in blue QLEDs. As both InP-based and ZnSe-based QLEDs adopt a “p-i-n” sandwich design, the operation of the device involves electrons and holes being injected into the QD layer through charge transport layers, ultimately recombining within the QDs to emit light. Consequently, strategies to optimize charge balance can be similarly applied in both cases.

One aspect involves the HOMO level of PVK (−5.8 eV), which aligns favorably with QDs for hole injection, in contrast to TFB (−5.3 eV), as shown in Fig. 9(a). Nevertheless, TFB's superior hole transfer stability necessitates consideration when selecting the optimal material<sup>114</sup>. Simultaneously, reducing the electron mobility of the electron transport layer, as demonstrated in Fig. 9(b) and 9(c), has proven effective in balancing charge injection within QLEDs<sup>69,72</sup>. Beyond that, Chen's

group<sup>67</sup> has developed a top-emitting device structure, as shown in Fig. 9(d). By integrating transparent indium zinc oxide as both the top electrode and a phase-tuning layer, this design achieves a saturated blue light emission that closely aligns with the Rec.2020 color standard. Additionally, due to its remarkable external coupling efficiency and balanced charge transport, the ZnSeTe-based QLEDs for blue emission exhibit an impressive EQE of up to 18.16%.

## Challenges and solutions for long-lifetime QLED

Due to the distinctive benefits and inherent constraints of InP-based and ZnSe-based QDs, the diversification of QD core-shell engineering and QLED interface modification techniques enables the PLQY of QDs to nearly reach 100%, whereas the EQE of QLEDs remains around 20%. Moreover, high PLQY does not inherently guarantee



**Fig. 9** | Enhancements in charge balancing of blue ZnSe-based QLEDs. (a) Illustration of the structure and energy level alignment in a conventional QLED device. (b) Modification of ZnMgO NPs through additional Mg reaction. (c) Employment of Sn-doped ZnO to mitigate electron over-injection. (d) Demonstration of ZnSeTe-based blue top-emitting QLEDs. Figure reproduced with permission from: (a) ref.<sup>114</sup>, Royal Society of Chemistry; (b) ref.<sup>72</sup>, (c) ref.<sup>69</sup>, American Chemical Society. (d) ref.<sup>67</sup>, Springer Nature.

long-lifetime QLEDs. It is imperative to tackle the issue of charge injection imbalance and field-enhanced electron delocalization in Cd-free devices during operation, as these factors cause a decrease in brightness. Nevertheless, the primary challenge in commercializing QLEDs revolves around developing cadmium-free QDs with prolonged lifetimes for full-color, cadmium-free displays, which requires QDs to serve as the self-luminous layer to possess not only high PLQY but also to maintain steady brightness during operation.

Meanwhile, unlike their red and green QLEDs, blue QLEDs exhibit unique degradation mechanisms that are yet to be fully elucidated, primarily hindered by their markedly inferior lifetime. Firstly, their intrinsic properties, such as a deep valence band and large bandgap, lead to an elevated hole injection barrier and increased surface defects, which can reduce performance and PLQY. Secondly, structural differences in device design and material compatibility for blue QDs necessitate specific considerations to optimize carrier injection and transport. Thirdly, degradation mechanisms such as charge accumulation and leakage, non-radiative recombination, and interfacial stability issues are more prevalent in blue QLEDs, leading to reduced efficiency and stability. Lastly, achieving high efficiency and accurate chromaticity for blue QLEDs is particularly challenging, further highlighting their unique degradation challenges compared to red and green QLEDs. Consequently, we embark on an overview of the degradation mechanisms of QLEDs, along with strategies to improve their lifetime performance.

### The degradation mechanisms of InP-based QLEDs

Despite attempts to mitigate surface traps through ligand passivation and minimize lattice mismatch issues with core-shell structures, the inherent chemical reactivity of In and P is too strong to prevent oxygen-induced traps in InP QDs completely. In QLEDs, these traps hinder exciton generation and radiative recombination by capturing injected carriers. Additionally, they contribute to the formation of trapped excitons with excessively rapid nonradiative decay rates, thus posing a significant challenge to achieving long lifetimes. For instance, Du's group<sup>115</sup> has elucidated the distinct loss mechanisms of InP-based QLEDs under low and high bias voltages. With the increase in shell thickness, exciton quenching caused by surface traps and spontaneous energy transfer independent of the field are effectively suppressed. Con-

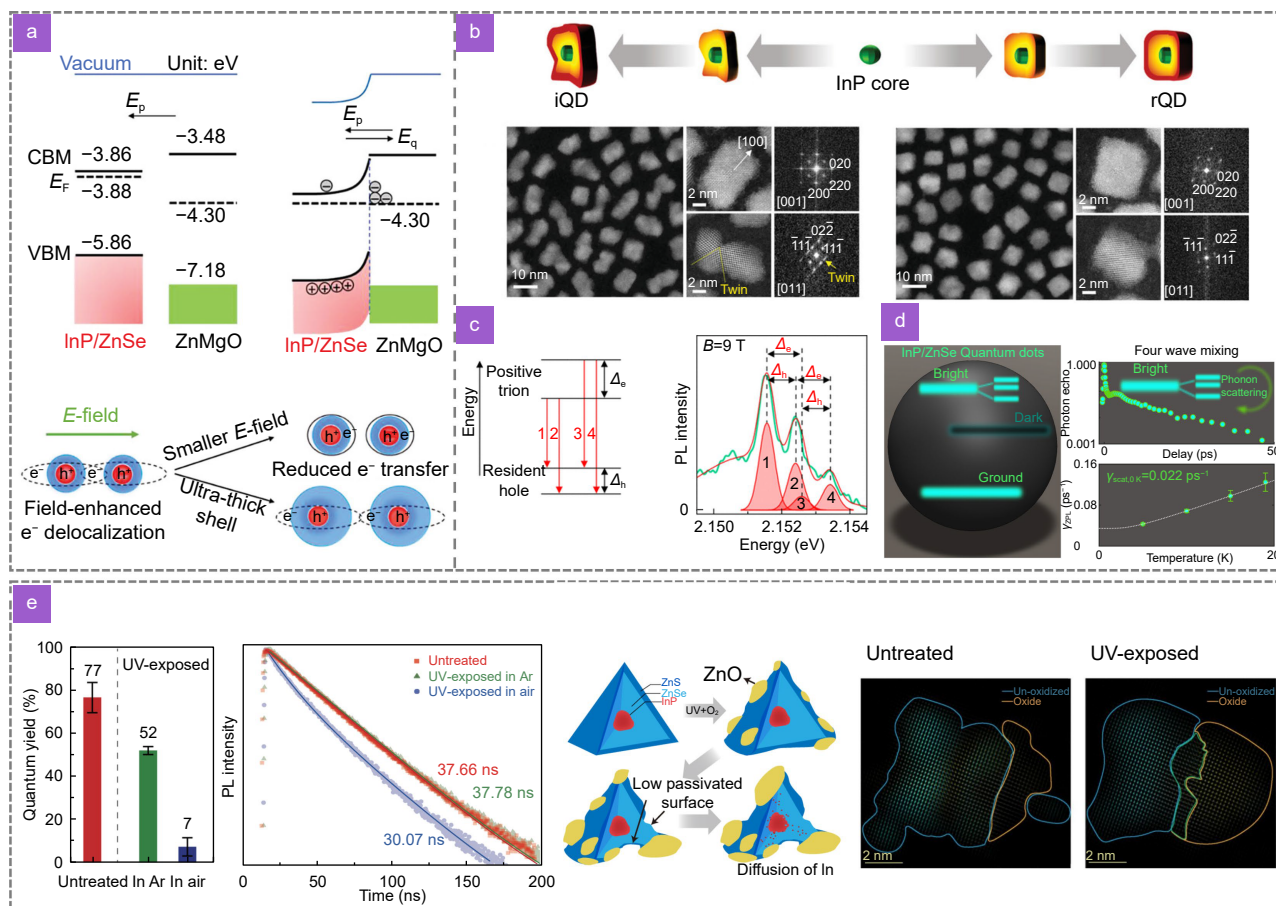
versely, the field-enhanced electron delocalization that leads to highly efficient energy transfer is one of the primary reasons for the sharp decline in EQE in InP-based QLEDs under high bias (Fig. 10(a)).

In addition, the challenges posed by the broad FWHM, size irregularity, and core-shell interfacial non-uniformity of InP QDs have been at the forefront of scientific inquiry. Kim et al.<sup>20</sup> have highlighted the strong dependence of charge carrier behavior in InP-QLEDs on the morphology of QDs, as depicted in Fig. 10(b). Compared to anisotropic QDs, isotropic counterparts exhibit significantly reduced hole trapping in InP/ZnSe/ZnS QDs and enhanced PLQY due to the effective elimination of stacking faults within the QDs. In anisotropic QDs, photo-generated charges are more prone to entering non-radiative recombination centers, and charge capture by defect sites occurs more readily. Remarkably, the difference in PLQY between the two types of QDs is not attributed to surface defects but rather to the extent of hole trapping.

Contrary to epitaxial III-V or colloidal II-VI QDs, phonon-mediated scattering among bright exciton states prevails as the primary dephasing mechanism in colloidal core-shell InP/ZnSe QDs. For example, Bayer et al.<sup>116</sup> provided evidence for the emission of positive triions through the analysis of Zeeman splitting into the spectral lines of individual InP/ZnSe/ZnS, as clearly illustrated in Fig. 10(c). By employing both one-photon and two-photon photoluminescence excitation on nanocrystal ensembles, the research conclusively designates the 1Sh state as the lowest energy hole level, in contrast to the 1Ph state. Moreover, individual InP/ZnSe QDs exhibit minimal spectral fluctuations, facilitating the observation of emission spectra with line widths limited by dephasing effects, as illustrated in Fig. 10(d)<sup>117</sup>.

Another issue is that the high susceptibility to oxidation of InP/ZnSe/ZnS QDs significantly impacts their emission efficiency, making emission quenching under the environment a persistent challenge. For instance, Park's team<sup>118</sup> observed that when individual InP/ZnSe/ZnS QDs are exposed to UV light, oxidation of the ZnS shell leads to the formation of ZnO, causing lattice mismatch with the host core lattice. This in turn creates dislocations and strains within the QD, prompting the diffusion of In atoms from the core to the surface. In particular, oxidative defects within the QD further contribute to incomplete passivation of the ZnSe layer, ultimately leading to the emergence of dangling bonds, as





**Fig. 10** | The degradation mechanisms of InP-based QLEDs. (a) The impacts of the electron delocalization and the associated energy transfers on the characteristics of InP-based QLEDs. (b) The STEM and HR-TEM images. (c) Photoluminescence spectra of QDs in varying magnetic fields. (d) Depiction of exciton decoherence as a phonon-mediated, thermally activated process. (e) Emission quenching of InP/ZnSe/ZnS QDs under UV exposure. Figure reproduced with permission from: (a) ref.<sup>115</sup>, (b) ref.<sup>20</sup>, John Wiley and Sons; (c) ref.<sup>116</sup>, American Chemical Society; (d) ref.<sup>117</sup>, American Chemical Society; (e) ref.<sup>118</sup>, Springer Nature.

demonstrated in Fig. 10(e).

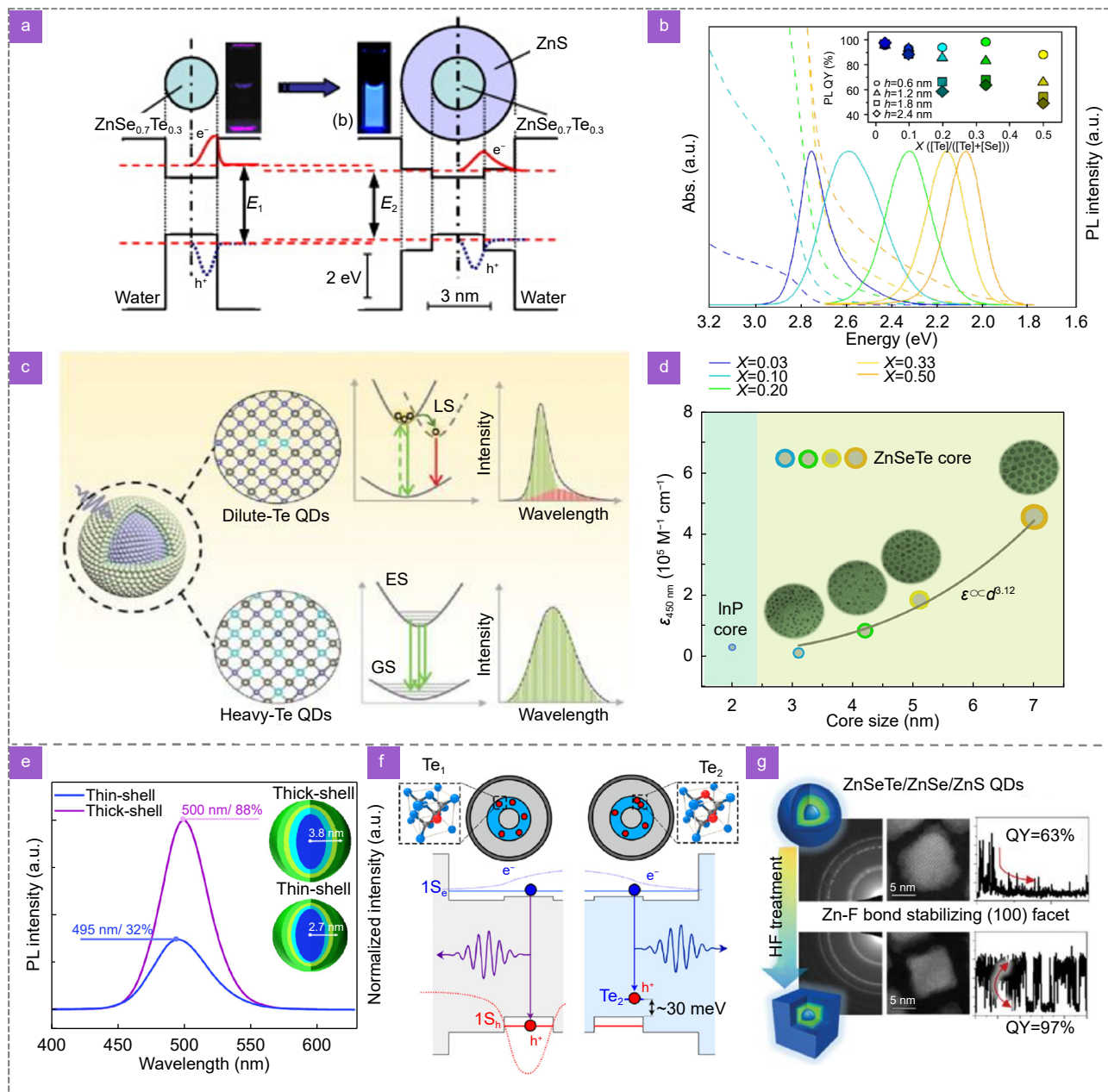
Recent developments in cadmium-free QD-LEDs have been significant and promising. The research team, comprising Professor Shen, Professor Feng, and Professor Tang, used their custom-developed electrically excited transient absorption technology to analyze the electronic transport characteristics of QLEDs<sup>119</sup>. Their study revealed that the ZnSeS intermediate layer hindered the efficiency of green InP-QLEDs. By experimentally and theoretically proving that a thicker ZnSe layer could replace ZnSeS to improve electron injection and reduce leakage, they achieved a peak EQE of 26.68% and a  $T_{95}$  lifetime of 1,241 hours for green InP-based QLEDs with an initial brightness of 1000 cd/m<sup>2</sup> at 543 nm.

### Te homogenization within ZnSe-based QLEDs

By taking advantage of the ZnS bandgap, the influence of the quantum confinement effect can be effectively miti-

gated through a thick shell that confines the electrons within the particles and thereby improve the PL efficiency and stability against changes in the pH of the solution (Fig. 11(a))<sup>120</sup>. Meanwhile, the integration of ZnTe with a narrower bandgap and ZnSe with a broader one allows for the tuning of the bandgap, which can be tailored to produce blue-emitting ZnTeSe QDs, as shown in Fig. 11(b)<sup>121</sup>. Moreover, the prevention of blue light leakage from light-emitting chips significantly relies on the absorption efficiency of QDs under blue excitation. Importantly, both the emission spectra and absorption capacity are exquisitely sensitive to changes in core size. Yang et al.<sup>58</sup> revealed that by evaluating the size-dependent molar absorption coefficients at 450 nm for a range of ZnSeTe cores and producing green-emitting ZnSeTe cores of varying sizes while preserving a uniform Se/Te ratio, as shown in Fig. 11(d).

In particular, the challenges in achieving high color



**Fig. 11** | Mechanism for the blue emission of ZnSe-based QLED. (a) Radial probabilities for the presence of electrons and holes of excitons in QD with/without irradiation. (b) Absorption (dashed line) and PL spectra (solid line) of ZnSe QDs with varying Te ratios and ZnSe shell thicknesses. (c) The temperature-dependent PL spectroscopy of the main and tail emission. (d) Size-dependent molar absorption coefficients. (e) PL spectra along with schematic diagrams. (f) Impact of nearest-neighbor pairs of Te atoms on the optical properties of ZnSe/ZnSe<sub>1-x</sub>Te<sub>x</sub>/ZnSe/ZnS NCs; Copyright 2022, American Chemical Society. (g) Morphological and crystalline structural changes upon excess HF treatment over the synthesis process. Figure reproduced with permission from: (a) ref.<sup>120</sup>, American Chemical Society; (b) ref.<sup>121</sup>, authors; ref.<sup>122</sup>, John Wiley and Sons; (c) ref.<sup>58</sup>, (e) ref.<sup>123</sup>, (f) ref.<sup>124</sup>, American Chemical Society; (g) ref.<sup>125</sup>, John Wiley and Sons.

purity blue QLEDs lie in the low-energy tail emission and spectral broadening observed in PL spectra. Wang et al.<sup>122</sup> found that with low Te doping ( $\text{Te/Se} < 20\%$ ), the asymmetric low-energy tail arises from localized state recombination by Te clusters, with hot carrier localization occurring within  $\sim 500$  fs. Above 20% Te/Se, the decrease

in electronegativity contrast results in a homogeneous charge density distribution, leading to carrier delocalization and the elimination of tail emission. The broadening of emission FWHM primarily stems from inhomogeneous broadening effects, as seen in Fig. 11(c). Moreover, the typically poor PLQY in ZnSeTe QDs is due to

ultrafast hot carrier/band-edge carrier trapping. In thick-shell samples, rapid decay is suppressed, while the slow component relates to radiative exciton recombination of long lifetimes. Effective passivation of band-edge electron traps in thicker shells minimizes photoinduced absorption from electron trapping states, as illustrated in Fig. 11(e)<sup>123</sup>.

Recently, Bae's group<sup>124</sup> has illuminated the morphological inhomogeneity within  $\text{ZnSe}_{1-x}\text{Te}_x$  alloy layers, specifically the presence of nearest-neighbor Te pairs, rather than size or compositional inhomogeneity, is the primary cause of dispersion observed in emission spectra and decay kinetics (Fig. 11(f)). Due to the difference in electronegativity between Se and Te, nearest-neighbor Te pairs in  $\text{ZnSe}_{1-x}\text{Te}_x$  alloys create localized hole states, leading to the formation of spatially separated excitons (delocalized electrons and localized holes in traps), which explains the inhomogeneous and homogeneous FWHM broadening with delayed recombination dynamics. In addition, HF treatment is a well-established method to remove dangling bonds and stacking faults in QDs. As illustrated in Fig. 11(g), Kim et al.<sup>125</sup> demonstrated that the addition of HF promotes the formation of Zn-F bonds, which in turn reduces surface energy and favors the preferential growth of (100) facets. This not only enhances the lattice order but also improves the PL emission properties. Additionally, an increase in HF concentration results in a more stable PL blinking behavior and a decrease in spectral diffusion.

## Recent advancements in color patterning

The advancement in QLED technology, featuring heightened efficiency and stability, is fostering a highly promising future for large-area display applications. Nevertheless, the endeavor to achieve high-resolution pixel arrays through the patterning of QD layers remains a formidable obstacle. An ideal QD patterning approach should embody high resolution, impeccable uniformity, minimal degradation of QDs, cost-effectiveness, and other favorable characteristics. At present, the development of full-color display solutions based on cadmium-free QDs is in its nascent stages, a topic we will explore further in subsequent sections.

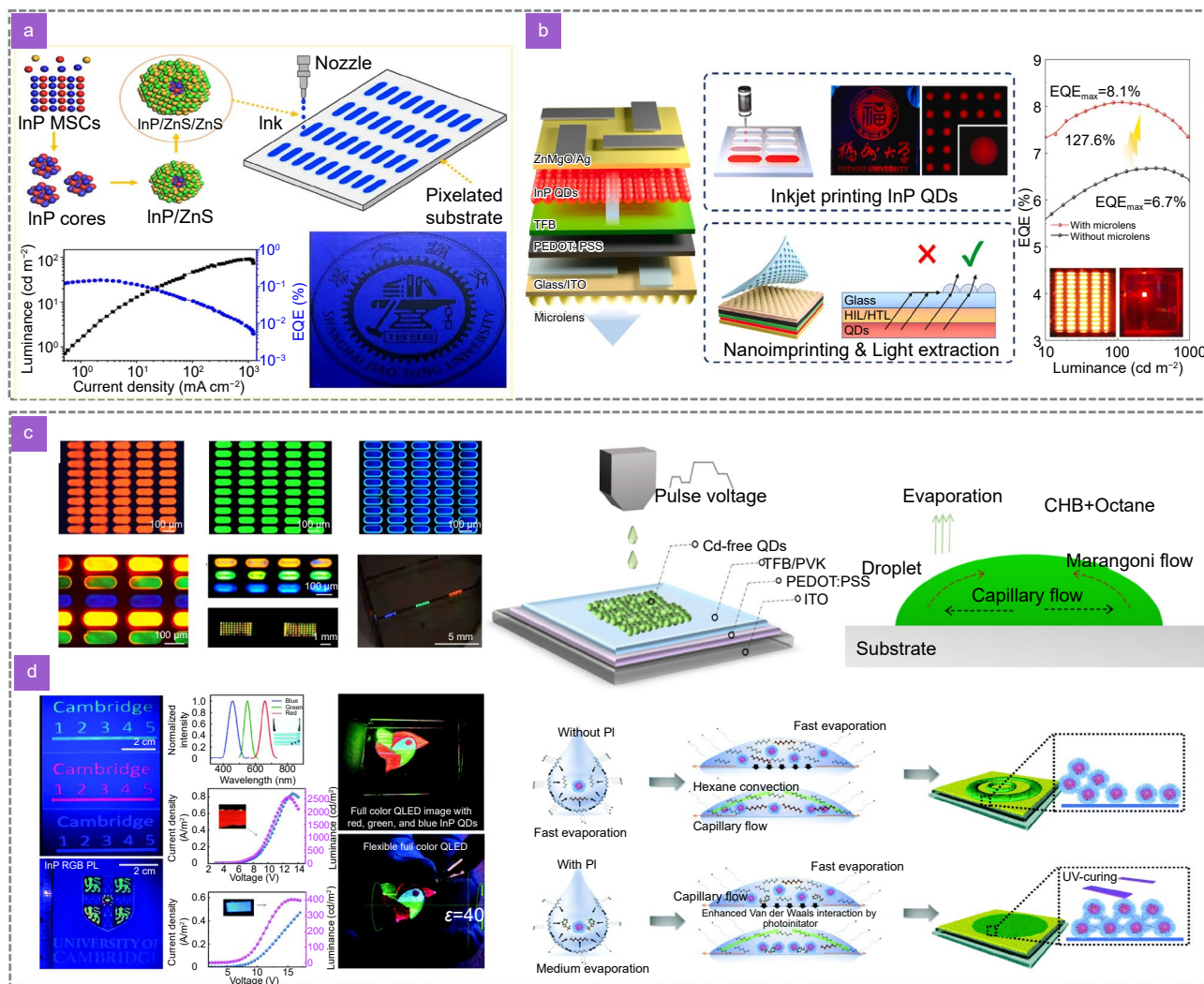
Typically, QLEDs produced via spin-coating methods require an inert atmosphere for fabrication, whereas the inkjet printing technique operates optimally under ambient conditions. However, eco-friendly InP QLEDs, un-

like their stable Cd-based counterparts, encounter challenges during production under ambient conditions, primarily due to the degradation of QD. In 2022, Li et al.<sup>126</sup> reported the first efficient and high-color-purity inkjet-printed QLED using blue InP/ZnS/ZnS QDs as the emission layer. A two-step heating and thick shell strategy was utilized to prepare large-sized (10.6 nm) blue InP/ZnS/ZnS QDs. The substantial ZnS shell layer effectively suppresses non-radiative FRET among densely packed QDs, leading to high PLQY in InP/ZnS/ZnS QD films. However, the pixelated blue InP QLED achieved a maximum brightness of 91  $\text{cd}\cdot\text{m}^{-2}$  and an EQE of 0.15%, as shown in Fig. 12(a).

Recent innovations in multi-component QD ink formulation have led to the development of inks with superior stability and printability, suitable for inkjet printing of InP QLED arrays. By carefully managing the dynamic interplay between capillary and Marangoni flows during the printing process, it is possible to achieve high-quality QD films on diverse substrates, including those featuring raised banks. Furthermore, the integration of ZnO microlens arrays, fabricated by nanoimprinting, has significantly improved the light extraction efficiency, leading to a maximum luminance of 17759  $\text{cd}\cdot\text{m}^{-2}$ , an EQE of 8.1%, and a current efficiency of 11.1  $\text{cd}/\text{A}$  (Fig. 12(b))<sup>127</sup>. These advancements are poised to accelerate the adoption of inkjet-printed, environmentally friendly QLEDs.

In addition, mitigating solubility issues of the underlying hole transport layer during inkjet printing is essential for achieving high pixel resolution and uniformity. For example, Kim et al.<sup>128</sup> employed a dual HTL layer consisting of TFB and PVK, effectively mitigating QD ink-induced HTL corrosion while preserving hole transport efficiency. Utilizing InP/ZnSeS red and green QDs along with ZnTeSe/ZnSe/ZnS blue QDs, a cadmium-free RGB inkjet-printed QLED was realized. Based on photolithography, high-resolution RGB color QLED pixels with dimensions of  $60\ \mu\text{m}\times 160\ \mu\text{m}$  were presented, as illustrated in Fig. 12(c). Furthermore, they employed hexane, which has a low boiling point, as the primary solvent, adding a small amount of octane (less than 10% by volume) to moderate the evaporation rate of the droplets. Upon depositing the QD ink containing the photoinitiator 2-hydroxy-2-methylpropiophenone onto the desired surface, the film underwent in-situ polymerization induced by the formation of radicals through ultraviolet irradiation (Fig. 12(d)). The crosslinking





**Fig. 12 |** Recent progress on eco-friendly QD patterning technologies. (a) Inkjet-printed QLED utilizing blue InP/ZnS/ZnS QDs as emission layer. (b) Nanoimprinting-aided inkjet-printed QLED for enhanced light extraction. (c) Cadmium-free RGB inkjet-printed QLED. (d) Comparison of InP-based green QLEDs printed with and without photoinitiator inclusion. Figure reproduced with permission from: (a) ref.<sup>126</sup>, (b) ref.<sup>127</sup>, Elsevier; (c) ref.<sup>128</sup>, John Wiley and Sons; (d) ref.<sup>129</sup>, Royal Society of Chemistry.

reaction among the ligands in the QDs reduced the inter-particle distances, leading to a flattened surface and enhanced environmental stability in the air. The maximum luminance of the printed InP-based green QLED reached 3600 cd·m<sup>-2</sup><sup>129</sup>.

## Summaries and perspectives

For two decades, QDs have sparked a revolution in displays owing to their distinctive luminescent properties and vivid color range. Researchers have diligently refined and innovated the materials and designs of QLEDs, propelling a remarkable surge in their capabilities. Currently, cadmium-free QLEDs in red, green, and blue QLED are nearly on par with Cd-QLEDs in terms of EQE. However, ensuring their stability poses the prima-

ry obstacle to achieving fully environmentally friendly, full-color displays. Furthermore, to expand the versatility of QLEDs into areas such as photomedicine, biosensing, lasers, and visual light communication, we encourage researchers to focus their efforts on the following directions.

i) Formulating precise and technical specifications. The methodologies employed by various laboratories for characterizing the same material exhibit similarities, yet there persists an absence of standardized definitions for QLED performance indicators, such as "device lifetime" and "device stability."

ii) Device performance of cadmium-free green QLEDs. So far, there is still a lot of room for improvement in the efficiency and lifespan of cadmium-free



QLEDs, especially for green QLEDs. The exchange of surface ligands for cadmium-free QDs (including InP and ZnSn types) represents an effective pathway toward achieving high-performance QLEDs. Possible effective strategies include crystal engineering and surface ligands with excellent optoelectronic stability for cadmium-free green QDs.

iii) Exploring the degradation mechanism of cadmium-free devices. Enhancing QLED stability requires a deep comprehension of their degradation mechanisms. It is essential to recognize that a high PLQY does not inherently guarantee a prolonged device life, due to the intricate relationship between QD electric capabilities, carrier transport layer stability, and QLED packaging technology. Possible effective strategies include reducing the sensitivity of QDs to electric fields, and material chemistry for high-performance HTLs.

iv) Exploring the patterning technologies. To meet the demands of ultra-high resolution, the patterning of the QD layer must adhere to high pixel density, uniformity, and low roughness. However, the stability of cadmium-free QDs remains a challenge, and full-color displays utilizing inkjet printing technology are still in their developmental stages. Therefore, it is crucial to explore and develop alternative patterning technologies that offer high resolution, low cost, and high yield for realizing full-color cadmium-free QLED displays.

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## Author contributions

Z. Y. supervised the work and made substantial revisions to the manuscript. P. G. wrote the draft of the manuscript. All authors discussed the results and reviewed the manuscript.

## Competing interests

The authors declare no competing financial interests.



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