Solution-processed green and blue quantum-dot light-emitting diodes with eliminated charge leakage

Yunzhou Deng,1,2,8, Feng Peng,2,8, Yao Lu,4,8, Xitong Zhu,1,8, Wangxiao Jin,1, Jing Qiu,4, Jiawei Dong,4, Yanlei Hao,1, Dawei Di,2, Yuan Gao,5, Tulai Sun,6, Ming Zhang,7, Feng Liu,1,7, Linjun Wang,4,4, Lei Ying,3, Fei Huang,8 and Yizheng Jin,1

Quantum-dot light-emitting diodes (QD-LEDs) promise a new generation of efficient, low-cost, large-area and flexible electroluminescent devices. However, the inferior performance of green and blue QD-LEDs compared with their red counterpart is hindering the commercialization of QD-LEDs in display and solid-state lighting applications. Here we demonstrate green and blue QD-LEDs with ~100% conversion of the injected charge carriers into emissive excitons. The key to success is the elimination of electron leakage at the organic/inorganic interface by using hole-transport polymers with simultaneous low electron affinity and reduced energetic disorder. Our devices exhibit high external quantum efficiencies over a wide range of luminance values (peak external quantum efficiencies of 28.7% for green and 21.9% for blue) and excellent stability (extrapolated $T_{95}$ lifetime is 580,000 h for green and 4,400 h for blue QD-LEDs). We expect our work to provide a general strategy for eliminating charge leakage in solution-processed LEDs featuring organic/inorganic interfaces.

Light-emitting diodes (LEDs) using solution-processable emitters, such as colloidal quantum dots (QDs)1–11, conjugated polymers,12–15 and metal halide perovskites16–20, are attractive alternatives to next-generation display and lighting technologies owing to their potential for the low-cost fabrication of efficient, large-area and flexible electroluminescent (EL) devices. Colloidal QDs are a unique class of solution-processable inorganic crystals featuring efficient, stable and high-colour-purity luminescence.11–25 CdSe-based QDs,25 and more recently, InP-based26 and ZnSe-based QDs,27,28 have been applied as the emissive materials in LEDs. Figure 1a shows a typical structure of the state-of-the-art QD-LEDs, which consists of QDs sandwiched between electron-transport layers based on ZnO nanoparticles and hole-transport layers (HTLs) based on polymers. To promote the performance of QD-LEDs, several effective strategies including advanced synthesis chemistry for enhancing photoluminescence quantum yields (PLQYs) of QDs19,21,24,27, band-structure tailoring of QDs for hole-injection improvement25,26, use of electrochemically stable ligands for eliminating in situ redox reactions29 and material design of ZnO electron-transport layers for achieving efficient electron injection and suppressed interfacial exciton quenching29,30, these efforts have enabled CdSe-based QDs to reach ~97% of the initial value of 100 cd m$^{-2}$, $T_{95}@100$ cd m$^{-2}$, >300,000 h, fulfilling the requirements of EL displays.30 However, EL efficiencies of the state-of-the-art green and blue QD-LEDs are still lower than the limits considering the near-unity PLQYs of QDs and the light out-coupling efficiencies of QD-LEDs. The operational lifetimes of green and blue QD-LEDs, electron leakage from the QDs to the HTLs is largely enhanced by both energetic disorder of organic HTLs and size discrepancy between the QDs and HTL segments. To suppress electron leakage, we modulated the HTL materials to simultaneously achieve a shallow lowest-unoccupied molecular orbital (LUMO) and reduced energetic disorder. This strategy finally results in ~100% conversions from charge carriers into excitons in QD-LEDs, leading to green and blue LEDs with overall performances surpassing those of previously reported solution-processed LEDs.

**Results**

Electron-leakage-induced efficiency loss in QD-LEDs. We investigate the discrepancies of efficiency losses in red-, green- and blue-coloured QD-LEDs with Cd-based QDs as emitters. Red (CdSe/CdZnSe/ZnS) core/shell/shell QDs, green (CdSe/CdZnSe/ZnS) core/shell/shell QDs and blue (CdZnSe/ZnS) core/shell QDs with similar diameters (~10 nm) and surface ligands (Supplementary Fig. 1) are incorporated into a unified QD-LED device operation. We reveal that in green and blue QD-LEDs, electron leakage from the QDs to the HTLs is largely enhanced by both energetic disorder of organic HTLs and size discrepancy between the QDs and HTL segments. To suppress electron leakage, we modulated the HTL materials to simultaneously achieve a shallow lowest-unoccupied molecular orbital (LUMO) and reduced energetic disorder. This strategy finally results in ~100% conversions from charge carriers into excitons in QD-LEDs, leading to green and blue LEDs with overall performances surpassing those of previously reported solution-processed LEDs.
structure (Supplementary Fig. 2a). Poly([9,9-diocytfluorenyl-2,7-diy]-co-(4,4’-(N-(4-sec-butylphenyl)diphenylamine)) (TFB), the benchmark hole-transport material for QD-LEDs, is employed as the HTL. All the devices show sub-bandgap turn-on characteristics (Fig. 1b), indicating the efficient injection of both electrons and holes into the QDs. Previous reports suggested that the sub-bandgap turn on of QD-LEDs may be a result of field-assisted or thermal-assisted charge injection. The internal quantum efficiency (IQE) of red QD-LEDs determined from the EQE and out-coupling efficiency (Supplementary Fig. 2) is close to the limit defined by the PLQY of the red QD film (Fig. 1c). In contrast, there are pronounced EL–PL efficiency gaps—discrepancies between the IQE of the EL devices and PLQYs of the QD films—for green and blue QD-LEDs (Fig. 1c).

Spectral characterizations indicate electron leakage into the HTLs in the green and blue QD-LEDs. The red QD-LED shows...
pure QD emission in the EL spectra (Fig. 1d). In contrast, parasitic emissions from TFB are observed in the EL spectra of green and blue QD-LEDs (Fig. 1d (arrows); Supplementary Fig. 3 shows the EL properties of TFB), suggesting exciton formation in the TFB. Transient PL characterizations (Supplementary Fig. 4) rule out the possibilities of energy-transfer processes from QDs to TFB or exciton dissociation induced by TFB. Hence, in the operation of blue and green QD-LEDs, exciton formation in the TFB suggests electron leakage, that is, electron transfer from the negatively charged QDs to the TFB HTL (QD $\rightarrow$ HTL $\rightarrow$ QD $+$ HTL$^-$). Given the low EL efficiency of TFB ($\sim$0.01%; Supplementary Fig. 3), the distinguishable parasitic emission (the intensity is almost three orders of magnitude lower than that of QD emission) in the EL spectra of green and blue QD-LEDs indicate non-negligible electron leakage, causing the EL-PL efficiency gaps. The leakage electrons can also be evidenced by the deterioration of TFB after the long-time operation of QD-LEDs$^{30}$, as shown by in situ PL measurements (Supplementary Fig. 5).

**Energetic-disorder-enhanced interfacial electron transfer.** It is of interest to understand the underlying mechanism enabling interfacial electron leakage. A glance at the energy-level diagram at the QD/HTL interface (Supplementary Fig. 6) would infer that the large energy offset between the LUMO of TFB and the conduction band (CB) of QDs ($\Delta E_{\text{LUMO,HTL-CB,QD}}$ $\approx$ $\sim$0.8 eV) makes the electron transfer from QDs to TFB extremely inefficient, if not impossible. We consider that this conventional picture is oversimplified because the interfacial charge-transfer rate also relies on the densities of the initial and final electronic states involved in the process. Given the uniqueness of the interface between crystalline inorganic QDs and amorphous organic HTLs and size discrepancy between the QDs and HTL segments. For polymeric HTLs, both static disorder (denoted by $\sigma$, the Gaussian standard derivation of density-of-states (DOS) distribution in the frontier orbitals) and dynamic disorder (denoted by $\lambda$, the reorganization energy) contribute to energetic disorder near the LUMO (Fig. 1c). The former is due to structural disorder (for example, variations in molecular packing and conjugation lengths of the segments) or defects$^{38-40}$, and the latter originates from strong electron–phonon interactions$^{39,40}$. Static disorder (broadened LUMO DOS) of the HTL causes more distributions of deep tail states, which are energetically favourable for electron leakage. Dynamic disorder of the HTL also facilitates electron transfer because the structural reorganization of the HTL reduces the average final-state energy and the energy fluctuation increases the distribution of energy levels with smaller energy barriers, both increasing the strength of effective electronic coupling between the HTL and QDs. Furthermore, a QD crystal (diameter, $\sim$10 nm) is considerably larger than an HTL segment (size, $\sim$1–2 nm). Therefore, the electron in a QD donor can be transferred into multiple HTL acceptors (Fig. 1a).

We carry out mixed quantum-classical simulations of the electron-transfer dynamics in one-dimensional (1D), two-dimensional (2D) and three-dimensional (3D) interfacial models (Fig. 1f), with the state-of-the-art crossing-corrected global flux surface hopping method implemented in a home-made SPADE simulation package (Methods and Supplementary Note 1$^{41-44}$). The effects of $\sigma$, $\lambda$, and $\Delta E_{\text{LUMO,HTL-CB,QD}}$ are considered. Our theoretical investigation suggests a unique mechanism for electron transfer at the QD/HTL interface, distinctive from those at the inorganic/inorganic or organic/organic interfaces. Namely, the dynamics are generally initialized by several incoherent electron hops or an indirect super-exchange process, driven by energetic disorders of polymer HTLs as well as entropy gain originating from the different numbers of QD donors and HTL acceptors. The transferred electron is further relaxed by the strong structural reorganization in the HTLs, thus preventing the back transfer of the stabilized electron from the HTLs to rigid QDs (Supplementary Fig. 7 shows the representative trajectories).

The simulation results (Fig. 1g and Supplementary Fig. 8) show that in the absence of static disorder and size difference between QDs and HTLs (grey-open triangles), the electron-transfer probability for a small $\lambda$ across a large energy barrier ($\Delta E_{\text{LUMO,HTL-CB,QD}}$ $\sim$ $\sim$0.5 eV) is negligible ($<0.02\%$), which is consistent with conventional understanding. With the increased $\lambda$ of the HTLs, the electron transfer from QDs to HTLs for a given $\Delta E_{\text{LUMO,HTL-CB,QD}}$ is enhanced in the 1D model (Fig. 1g and Supplementary Fig. 8, grey-open circles). Notably, when the static energetic disorder of the HTL segments is introduced, the electron-transfer probability is enhanced by several orders of magnitude for a large $\Delta E_{\text{LUMO,HTL-CB,QD}}$ of over $\sim$0.5 eV (Fig. 1g and Supplementary Fig. 8, grey-solid circles). Moreover, the results of the 2D and 3D configurations indicate a further increased electron-transfer probability (Fig. 1g and Supplementary Fig. 8, red and blue solid circles). Overall, the results predict a non-negligible electron-transfer probability despite a seemingly huge $\Delta E_{\text{LUMO,HTL-CB,QD}}$ and support the existence of the electron-leakage channels in blue and green QD-LEDs (Fig. 1c). Regarding red QD-LEDs, the absence of such an electron-leakage channel is reasonable because of the relatively deep CBs of red QDs and hence a considerably large $\Delta E_{\text{LUMO,HTL-CB,QD}}$ of $\sim$1.4 eV (Supplementary Fig. 6).

HTL with shallower LUMO and reduced energetic disorders. These findings motivate us to propose a new design principle for the QD/HTL interface to boost the performance of green and blue QD-LEDs. As shown in Fig. 2a, a conventional strategy is to enhance hole injection$^{45}$ for more efficient exciton formation (QD $\rightarrow$ HTL$^-$ $\rightarrow$ QD$^+$ $+$ HTL) (ref. 14). Here we propose to use HTLs with shallower LUMO and reduced energetic disorders to suppress electron leakage (QD $\rightarrow$ HTL $\rightarrow$ QD $+$ HTL$^-$).

Our new principle requires the molecular structure of HTLs to simultaneously possess rigid backbones and limited conjugation. Accordingly, we employ a co-polymer of poly((9,9-dioc-tylyfluorenyl-2,7-diyl)-alt-(9-(2-ethylhexyl)-carbazole-3,6-diyl)) (PF8Cz). Compared with the benchmark HTL of TFB (Fig. 2b, left), the non-planar, propeller-like triphenylamine unit$^{46}$ is replaced by a planar 3,6-carbazole unit (Fig. 2b, right) in PF8Cz, providing a more rigid framework and presumably more ordered molecular packing that reduces the energetic disorder. For the 3,6-carbazole-based co-polymer, the nitrogen atom possesses strong electron-donating properties and meanwhile breaks the conjugation$^{46,47}$. Figure 2b shows the theoretically optimized geometries of PF8Cz and TFB dimers (Supplementary Fig. 9). Density functional theory (DFT) calculations demonstrate considerably smaller reorganization energy of PF8Cz ($\Delta = 0.32$ eV for monomers and 0.21 eV for dimers) than TFB ($\Delta = 0.47$ eV for monomers and 0.28 eV for dimers), indicating reduced electron–phonon interactions and dynamic disorder (Supplementary Fig. 10, Supplementary Table 1 and Supplementary Note 2). Furthermore, the more localized electron distribution on the fluorene unit in PF8Cz (Supplementary Fig. 9) offers a higher LUMO energy relative to that of TFB (by 0.24 eV).

The more ordered molecular packing in PF8Cz films than that in TFB films is verified by grazing-incidence wide-angle X-ray scattering (GIWAXS) characterizations. As shown in the 2D GIWAXS patterns (Fig. 2c), the lamellar-stacking features ($q_C = 0.3-0.4 \AA^{-1}$, indicated by arrows) of the TFB and PF8Cz films are observed. The scattering ring corresponding to lamellar stacking in the PF8Cz film is considerably sharper than that of the TFB film, which is also shown by the stronger and better-defined scattering peaks of PF8Cz in the line-cut profiles (Fig. 2d). The crystal coherence length ($L_C$) determined from the breadth of the fitted peak is used to evaluate the degree of molecular ordering in the HTLs$^{48}$. The $L_C$ value of lamellar stacking in the PF8Cz film (25.2 Å) is larger than
that in the TFB film (16.1 Å), suggesting a more ordered lamellar stacking in PF8Cz. These results indicate better structural order in the PF8Cz film, which contributes to the low energetic disorder of PF8Cz.

The desired electronic properties of PF8Cz (Fig. 2b) are validated by spectroscopic characterizations. Both absorption and PL spectra of the PF8Cz film are blueshifted compared with those of the TFB film (Fig. 2c). The optical bandgap of the PF8Cz film is
High-efficiency green and blue QD-LEDs based on PF8Cz HTLs. a, Current density-voltage-luminance characteristics of green (top) and blue (bottom) QD-LEDs. The devices exhibit sub-bandgap turn-on voltages. The photographs of the operating QD-LEDs are also shown (scale bar, 5 mm). b, EL spectra of green and blue QD-LEDs based on PF8Cz HTLs (solid lines) and TFB HTLs (dashed lines). The arrows indicate the decrease in parasitic emissions from HTLs in PF8Cz-based devices compared with TFB-based devices. c, Current efficiencies (left axis) and luminous efficiencies (right axis) of green and blue QD-LEDs. d, EQEs of green and blue QD-LEDs in the luminance range for display and general lighting applications. e, IQEs of green and blue QD-LEDs and the corresponding PLQYs of the QD films.

3.06 eV, larger than that of the TFB film (2.88 eV). Ultraviolet photoelectron spectroscopy (UPS) measurements indicate an identical highest-occupied molecular orbital (HOMO) onset energy (−5.4 eV) for both PF8Cz and TFB films (Fig. 2f). Accordingly, the LUMO onset of the PF8Cz film is estimated to be upshifted by ~0.2 eV compared with that of the TFB film (Fig. 2b), which favours the blocking of electron leakage.

Furthermore, the reduced energetic disorder of the PF8Cz film is verified by the DOS profiles and band-bending analyses. The widths of the high-kinetic-energy peaks of valence spectra correspond to the extent of DOS broadening caused by energetic disorder. According to Fig. 2f, Gaussian fitting obtains a HOMO DOS width (σ_{HOMO}) of 0.25 ± 0.02 eV for PF8Cz, narrower than that of TFB (0.37 ± 0.04 eV). This result is in line with our analyses on the band-bending profiles (Fig. 2g), that is, shifts in the surface work functions with increased thicknesses of PF8Cz or TFB films on conductive poly(3,4-ethylenedioxythiophene):polystyrenesulfonate (PEDOT:PSS) substrates. Given the similar HOMO levels of TFB and PF8Cz films, the degree of band bending, which is ascribed to spontaneous charge transfer from the substrates into the tail states of the polymers (Supplementary Fig. 11), can be correlated with the width of DOS distributions and thus the energetic disorder of the HTLs.

The smaller work-function shifts of the PF8Cz films on PEDOT:PSS than those of the TFB films on PEDOT:PSS (Fig. 2g) imply a narrower distribution of HOMO DOS and reduced energetic disorder of PF8Cz.

Control experiments show that the surface morphology, molecular packing features and widths of DOS distributions of the PF8Cz films are not significantly altered by using materials with different molecular weights and dispersion indices (Supplementary Fig. 12). Characterizations on the films deposited under different processing conditions also show that PF8Cz films possess more ordered molecular packing than TFB films (Supplementary Fig. 12). The results suggest an inherently lower energetic disorder of PF8Cz HTLs.

We evaluate the electron-blocking properties of HTLs by analysing the band-bending characteristics of HTL films deposited onto low-work-function substrates of Sm, which results from the spontaneous electron transfer from Sm to HTLs (Supplementary Fig. 13). The films on Sm show pronounced band bending, whereas the surface work functions of the PF8Cz films on Sm are almost constant (Fig. 2h). The results verify that the shallower LUMO and reduced energetic disorder of PF8Cz than those of TFB lead to superior electron-blocking properties of PF8Cz.

High-performance QD-LEDs based on PF8Cz HTLs. Finally, we show that by using PF8Cz as the HTL, substantial improvements in EL performance are achieved in both green and blue QD-LEDs. The devices exhibit sub-bandgap turn-on voltages (Fig. 3a), similar to previously reported green and blue QD-LEDs with efficient charge injection. This feature implies that the combination of PF8Cz HTLs with a deep HOMO level (approximately equal to that of TFB) and Se-rich QDs can achieve efficient hole injection in QD-LEDs. The parasitic emissions of HTLs are substantially suppressed in PF8Cz-based QD-LEDs compared with those in TFB-based devices (Fig. 3b; Supplementary Fig. 14a shows the EL properties of PF8Cz). This feature validates the desirable electron-blocking properties of PF8Cz. Consequently, PF8Cz-based QD-LEDs demonstrate unprecedentedly high efficiencies. A luminous efficiency of 162 lm W⁻¹ and a current efficiency of 127 cd A⁻¹ are realized for the green device (Fig. 3c). The green and blue QD-LEDs show peak EQEs of 28.7% and 21.9%, representing the highest values for green and blue QD-LEDs to the best of our knowledge, respectively.
of ~580,000 h or a PF8Cz-based green and blue QD-LEDs. The 95 lifetime at an applications (Fig. 3d).

The luminance required for flat-panel display and general lighting solution-processed LEDs (Supplementary Fig. 16) and readily cover These wide high-efficiency windows are exceptional among previously reported solution-processed LEDs, including QD-LEDs, all performance of our green and blue devices is higher than those QD-LEDs (Figs. 1b and 3a) show no evidence of enhanced hole transport in PF8Cz-based QD-LEDs. The results further validate that the efficiency improvement is primarily due to the enhanced electron-blocking properties of PF8Cz. Impressively, green QD-LEDs show EQEs of >20% for luminance across four orders of magnitude (20 to over 200,000 cd m\(^{-2}\)), and blue QD-LEDs maintain EQEs of >20% in a wide luminance range of 400–25,000 cd m\(^{-2}\). These high-efficiency windows are exceptional among solution-processed LEDs (Supplementary Fig. 16) and readily cover the luminance required for flat-panel display and general lighting applications (Fig. 3d).

We highlight that long operational lifetimes are achieved in PF8Cz-based green and blue QD-LEDs. The \(T_{95}\) lifetime at an initial luminance of 11,220 cd m\(^{-2}\) \((T_{95}@11,220\,\text{cd}\,\text{m}^{-2})\) of a green device is determined to be 71 h, corresponding to a \(T_{95}@100\,\text{cd}\,\text{m}^{-2}\) of ~580,000 h or a \(T_{95}@1,000\,\text{cd}\,\text{m}^{-2}\) of ~7,200 h (Fig. 4a and Supplementary Fig. 15). This unprecedented lifetime of the green device sufficiently exceeds the requirements for EL displays. The blue QD-LED realizes a \(T_{95}\) lifetime of 44 h at an initial luminance of 1,150 cd m\(^{-2}\), indicating a record-long \(T_{95}@100\,\text{cd}\,\text{m}^{-2}\) of ~4,400 h or \(T_{95}@1,000\,\text{cd}\,\text{m}^{-2}\) of ~57 h (Fig. 4b and Supplementary Fig. 15). The \(T_{95}\) lifetimes (the time when the luminance decays to 50% of the initial value) at an initial luminance of 100 cd m\(^{-2}\) \((T_{95}@100\,\text{cd}\,\text{m}^{-2})\) are estimated to ~2,570,000 h for the green QD-LED and ~24,000 h for the blue QD-LED (Supplementary Fig. 15). According to a comparison of several key metrics (Supplementary Table 2), the overall performance of our green and blue devices is higher than those previously reported solution-processed LEDs, including QD-LEDs, organic LEDs and perovskite LEDs.

Comparative studies were conducted on a random co-polymer with the backbone consisting of 2,7-fluorene and 3,6-carbazole (denoted as R-PF8Cz), in which additional energetic disorder is introduced by random variations in electronic structures of the conjugation units. The results (Supplementary Fig. 17) confirm the general and critical impacts of energetic disorder and electron-blocking properties of HTLs on the device performance.

Conclusion

We have identified that electron leakage from QDs to HTLs, which is greatly enhanced by the energetic disorder of polymeric HTLs and geometric factors of the interfacial materials, is a major efficiency-loss channel in green and blue QD-LEDs. Accordingly, we offer a new design principle of employing HTLs with a shallower LUMO level and reduced energetic disorder to eliminate electron leakage. Advances in the fundamental understanding close the long-existing EL–PL efficiency gap for green and blue QD-LEDs, resulting in ~100% conversion of the injected charge carriers into emissive excitons. Our devices demonstrate exceptionally high efficiencies in wide ranges of luminance and record-long operational lifetimes, representing the best-performing solution-processed green and blue LEDs. Future efforts on molecular design and synthesis strategies of carbazole-based polymers\(^2\) could enhance the conductivity and electrochemical stability of HTLs, which shall further improve the power-conversion efficiencies and operational lifetimes of green and blue QD-LEDs. Moreover, improving the electrochemical stability of blue QDs and ZnO-based electron-transport materials might also be critical to boost the operational lifetimes of blue QD-LEDs towards practical applications. Our approach for the elimination of charge-leakage channels may inspire the design of other solution-processed LEDs with organic/inorganic interfaces.

Online content

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Fig. 4 | Operational lifetimes of QD-LEDs based on PF8Cz HTLs. a,b, Luminance as a function of operational time for green (a) and blue (b) QD-LED. The lifetimes \((T_{95})\) at various initial luminance \((L_{in})\) values are shown in the insets. The acceleration factors \((n)\) are fitted according to the empirical relationship of \(\frac{L_{in}}{T_{95}}\) = constant.
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QD-LEDs were bias stressed at a constant current density of 10 mA cm$^{-2}$ using a commercial LED-lifetime test system (Guangzhou Crysco Equipment). The operational lifetimes of QD-LEDs were measured under ambient conditions by monitoring the photometric quantities. The lamp (HL-3 plus, Ocean Optics) was used to determine the spectral luminous efficiency. The standard spectral luminous efficiency was calibrated by using a NIST-traceable radiant-flux standard (Ocean Optics) coupled to a spectrometer (QE Pro, Ocean Optics). The absolute radiometric characteristics and EL spectra were measured with a home-made system consisting of a digital source meter (Keithley 2400) and an integrating sphere (FOIS, Cary 5000). The current density–voltage–luminance characteristics were measured with a predefined value (20 ns for the 1D and 2D models and 4 ns for the 3D models) of electron cut-off. The binding energies (binding) were calculated by using theelectron cut-off. The binding energies (binding) were calculated by using the

UPS characterization and analyses. The UPS measurements were conducted with a Thermo Scientific ESCALAB 250Xi electron source operated in a high vacuum (radiation source, He i at 21.2 eV). The minimum kinetic energy (up to 20 eV) of the UPS spectrum was determined from the secondary electron cut-off. The binding energies (binding) were calculated by using the relationship $E_{\text{binding}} = E_{\text{Fermi}} - E_{\text{peak}}$. Then, the values measured were obtained by shifting the binding-energy scale to the energy scale relative to vacuum by $E = E_{\text{binding}} + E_{\text{Fermi}}$. The DOS widths were obtained according to an established method in the literature. The overall widths, in terms of Gaussian standard deviations, were determined by Gaussian fitting of the low-energy edges. The UPS Spectra were measured with a Nicolet iS50 Spectrometer (Thermo Fisher Scientific). The DOS widths ($\sigma_{\text{INS}}$) were determined by Gaussian fitting of the low-energy edges. The crystal coherence lengths were determined from the full-width at half-maximum of the diffraction peaks according to the Scherrer equation.

Other characterizations. The thickness of the films were characterized with a step profiler (P-8 Stylus, KLA Tencor). The cross-sectional sample of the QD-LED was prepared using a dual-beam focused ion beam system (Quadra 3D, FEI). The transmission electron microscopy images, high-angle annular dark-field scanning transmission electron microscopy images, and energy-dispersive X-ray spectrometry electron mapping results were obtained with a Cs aberration-corrected FEI Titan G2 80–200 ChemiSTEM instrument. The Fourier-transform infrared spectra were measured with a Nicolet 6700 Spectrometer (Thermo Fisher Scientific). The solid-state nuclear magnetic resonance (NMR) characterizations were performed on a Bruker Avance 600 III HD spectrometer (magnetic-field strength, 14.1 T) at the resonance frequency of 599.8 MHz for $^1$H.
DFT calculations. The DFT calculations of TFB and PF8Cz monomers and dimers were carried out at the B3LYP/6-31G* level with the Gaussian 16 package. A small QD, namely, Cd$_{68}$Se$_{55}$Cl$_{26}$, was studied at the B3LYP/LANL2DZ level. The reorganization energy was calculated as $\lambda = E(Q') - E(Q) + E'(Q') - E'(Q)$, where $E$ and $E'$ represent the energies of neutral and negatively charged molecules, whereas $Q$ and $Q'$ indicate the optimized geometric configurations corresponding to the neutral and negatively charged molecules, respectively. More details are provided in Supplementary Note 2.

Data availability
All data that support the plots within this paper and other findings of this study are available from the corresponding authors upon reasonable request.

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Author contributions
Y.J., together with F.H. and L.W., conceived the idea and supervised the work. Y.D. fabricated the high-performance QD-LEDs, conducted the spectral and electrical characterizations, carried out the optical modelling, and analysed the results under Y.J’s supervision. F.P. synthesized the high-quality PF8Cz and assisted in analysing the structural properties under the supervision of F.H. and L.Y. L.W. developed the charge-transfer simulation models and performed the theoretical analysis. Under the supervision of L.W., J.D. and J.Q. wrote the main codes of the SPADe software. J.Q. calculated the energy levels of QDs under the effective mass approximation, and Y.L. performed all the DFT calculations and dynamics simulations of the interfacial electron leakage. X.Z. assisted the device fabrication, conducted the optical characterizations, carried out the Kelvin probe measurements and analysed the hole-only devices under Y.J’s supervision. W.J. assisted in the characterizations of QDs and QD-LEDs. Y.H. assisted in the fabrication of QD-LEDs. Y.G. assisted in the characterization of CdSe-based QDs. T.S. conducted the high-angle annular dark-field scanning transmission electron microscopy and energy-dispersive X-ray spectroscopy characterizations. M.Z. and F.L. conducted the GIWAXS experiments and analysis. D.D. participated in data analysis and provided major revisions. All the authors discussed the results and commented on the manuscript.

Competing interests
The authors declare no competing interests.

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Correspondence and requests for materials should be addressed to Linjun Wang, Lei Ying, Fei Huang or Yizheng Jin.

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