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Guogang Li

ggli@cug.edu.cn

China University of Geosciences <https://orcid.org/0000-0002-0523-5621>

Wanying Zhang

Wuhan University of Technology

Mengjie Li

China University of Geosciences

Yiran Zhao

China University of Geosciences

Xuanyang He

China University of Geosciences

Tongtong Wang

China University of Geosciences

Xinyang Zhang

China University of Geosciences

Yingwei Wang

Central South University <https://orcid.org/0000-0003-1828-8420>

Fan Xia

State Key Laboratory of Geomicrobiology and Environmental Changes, Faculty of Materials Science and Chemistry, China University of Geosciences <https://orcid.org/0000-0001-7705-4638>

Keqiang Chen

China University of Geosciences <https://orcid.org/0000-0002-5741-1236>

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Dual-site surface sulfidation enables efficient CsPbI₃ quantum dot light-emitting diodes with ultralow roll-off

Wanying Zhang^{1,2}, Mengjie Li¹, Yiran Zhao¹, Xuanyang He¹, Tongtong Wang¹,
Xinyang Zhang¹, Yingwei Wang³, Fan Xia^{1,4}, Keqiang Chen^{1,2,*}, Guogang Li^{1,2,*}

¹Faculty of Materials Science and Chemistry, China University of Geosciences, Wuhan 430074, P. R. China

²Shenzhen Research Institute, China University of Geosciences, Shenzhen 518052, P. R. China

³Hunan Key Laboratory of Nanophotonics and Devices, School of Physics and Electronics, Central South University, Changsha 410083, P. R. China

⁴State key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, 388 Lumo Road, Wuhan 430074, P. R. China

Correspondent:

[*] Prof. Keqiang Chen

E-mail: chenkeqiang@cug.edu.cn

Prof. Guogang Li

E-mail: ggli@cug.edu.cn

Abstract

Metal halide perovskite quantum dots (QDs) are promising materials for developing low-cost and high-performance quantum dot-based light-emitting diodes (QLEDs). Despite external quantum efficiency (EQE) comparable to II–VI/III–V QLEDs, perovskite QD counterparts achieve peak EQE at low luminance/current density and exhibit significantly more pronounced efficiency roll-off, owing primarily to their ionic character and soft lattice. Engineering a robust surface is essential to ensure the structural integrity of perovskite QDs, yet achieving this remains a critical challenge. Here, we proposed a complete surface sulfidation strategy mediated by $(\text{H}_2\text{S})_2$ dimer that can simultaneously occupy both A-site and X-site of CsPbI_3 QDs, the dual-site passivation strategy leads to constructing a robust surface. Benefiting from the reinforced structural integrity, LEDs incorporating the treated CsPbI_3 QDs possess increased ion migration barrier, balanced charge injection, and suppressed Auger recombination. These combined improvements yield a peak EQE of 30.71% at 52 mA cm^{-2} and $1,089 \text{ cd m}^{-2}$, along with dramatically suppressed efficiency roll-off (maintaining EQEs of 18.59% at $\sim 500 \text{ mA cm}^{-2}$ and 25.01% at $\sim 5,000 \text{ cd m}^{-2}$) and a substantial improvement in operational half-lifetime.

Metal halide perovskite quantum dots (QDs) exhibit tunable emission across the entire visible region, combined with high photoluminescence quantum yields (PLQY) and narrow emission linewidths¹⁻⁵. These properties make them promising materials for developing low-cost quantum dot-based light-emitting diodes (QLEDs) that offer superior color saturation and exceptional quantum efficiency, positioning them as a leading candidate for next-generation display and lighting technologies⁶⁻⁸. Although perovskite QLEDs have achieved external quantum efficiency (EQE) comparable to that of conventional II–VI and III–V QD-based technologies, their EQEs peak at low luminance and exhibit significantly severer efficiency roll-off at high driving current density, posing a substantial obstacle to their practical application⁹⁻¹³.

A primary culprit behind this roll-off is nonradiative Auger recombination, whose rate scales with the cube of carrier density and is particularly pronounced under the quantum and dielectric confinement of QDs^{14,15}. Reducing the exciton binding energy (E_b) and dielectric confinement through high-dielectric-constant ligand passivation is an effective way to promote spatially homogeneous carrier distribution, thereby suppressing Auger recombination¹⁶. However, these improvements remain insufficient, perovskite QDs continue to face a fundamental constraint. Their ionic bonding and soft lattice, distinct from II–VI and III–V QDs with strong covalent bonding, make them particularly vulnerable to electric-field-driven ion migration and structural damage, leading to rising defect state density, aggravated Auger recombination, and severe efficiency roll-off¹⁷⁻²⁰. Engineering a robust surface is therefore imperative to ensure the structural integrity of perovskite QDs. Surface sulfidation is believed as a promising strategy to passivate perovskite films with robust surface due to the strong Pb–S bond and extremely low solubility product constant of PbS²¹. Nevertheless, the excessive strength of Pb–S bond, in turn, poses a fundamental constraint for perovskite QD passivation. Its uncontrolled formation can readily outcompete the relatively low internal lattice energy of perovskite, thereby providing a thermodynamic driving force for lattice disruption, PbS formation, PL quenching, and eventual structural degradation of the perovskite QDs²²⁻²⁴. Moreover, current sulfidation strategies are limited to X-site passivation, which are intrinsically incapable of stabilizing the weakly bound A-site

alkylamine ligands or mitigating their dielectric confinement effect. These failures directly lead to ligand desorption, which undermines structural integrity and, consequently, accelerates efficiency roll-off in LEDs^{25,26}.

In this work, we achieved a precisely controlled complete surface sulfidation of CsPbI₃ QDs utilizing hydrogen sulfide (H₂S), which involved a rapid X-site occupation via strong Pb–S bond formation, and followed by the gradual displacement of long-chain ligands at the A-site. The unique dual-site passivation process conferred a robust surface on CsPbI₃ QDs, resulting in a significantly reduced surface defect density, suppressed ion migration, balanced charge injection, and thus minimized Auger recombination. Consequently, surface sulfidation of QDs produced LEDs with dramatically enhanced performance, elevating the EQE from 6.11% to a 30.71% and extending the operational half-lifetime from 27 to 1331 min. Particularly, the optimized devices exhibited substantially suppressed efficiency roll-off, retaining an EQE of 18.59% and 25.01% at a high current density of $\sim 500 \text{ mA cm}^{-2}$ and high brightness of $\sim 5,000 \text{ cd m}^{-2}$, respectively, underscoring their exceptional potential for practical applications.

Dual-site surface sulfidation

We initiated the surface sulfidation by directly injecting H₂S/N₂ gas mixture into a sealed vial containing CsPbI₃ QDs solution (dispersed in octane), additional experimental details are described in the Materials and Methods section of the Supplementary Materials. We systematically examined the dosage-dependent effects of H₂S on CsPbI₃ QDs. Throughout the 0–2 mL range, their crystal phase remains stable, whereas their optical properties evolve significantly (Supplementary Fig. 1). The absorption intensity in the visible region increases progressively with H₂S addition (Supplementary Fig. 1b), correlated with a marked enhancement in the PLQY (Supplementary Fig. 1c). A slight blue-shift and a near-unity PLQY are achieved at the 1 mL H₂S optimum (Supplementary Fig. 1c), signifying effective surface defect passivation and a substantial decrease in defect state density. Over-sulfidation via further H₂S addition results in the formation of trace PbS and additional defects

(Supplementary Fig. 1d), which are directly manifested as emission redshift along with PLQY degradation (Supplementary Fig. 1e). Time-resolved PL (TRPL) measurements corroborate this interpretation, revealing a maximum PL lifetime of 57.3 ns at the 1 mL H₂S dosage (Supplementary Fig. 1f). Accordingly, this condition was established as optimal for subsequent QD passivation studies.

It should be pointed out that the surface sulfidation was conducted in the dark at 4 °C for 12 hours to ensure completion. It is noteworthy that the passivation time is crucial, as the samples treated under identical conditions but for longer durations exhibited a correlated increase in PL intensities and plateaued after 12 hours (Supplementary Fig. 2). Given that Pb–S bond formation is highly favorable both thermodynamically and kinetically, the slow passivation observed suggests that the overall process may be governed by a more complex mechanism. To this end, we conducted a comprehensive study on the surface chemical states of CsPbI₃ QDs, systematically investigating three distinct samples: the as-synthesized (pristine), those modified for 1 min (control) or 12 hours (treated). Fig. 1a demonstrates the Pb L₃-edge Fourier transformed k²-weighted R-space extended X-ray absorption fine structure (EXAFS) spectra of these QDs, characteristic peaks of Pb–I (3.22 Å) alongside Pb–S (2.36 Å) bonding can be distinctly found in both control and treated QDs, verifying the successful surface sulfidation at X-site. This finding is further corroborated by wavelet transform analysis (Supplementary Fig. 3), which shows a new intensity maximum specific to Pb–S scattering in these samples, in contrast to the pristine QDs. The successful incorporation of sulfur into CsPbI₃ QDs is also confirmed by transmission electron microscope–energy dispersive X-ray spectroscopy (TEM-EDX) mapping (Supplementary Figs. 4–6) and X-ray photoelectron spectroscopy (XPS, Supplementary Fig. 7). The Pb 4f core-level spectrum (Supplementary Fig. 7b) shifts to higher binding energy upon H₂S treatment, which should be contributed to the formation of strong Pb–S bonds at the X-site. Furthermore, the S 2p and N 1s peaks exhibit inverse trends in intensity with prolonged treatment, the former increases while the latter decreases. Consistently, Fourier-transform infrared (FTIR) mapping (monitored at 1,467 cm^{−1}, Figs. 1b–d), FTIR spectra (Supplementary Fig. 8a), and

nuclear magnetic resonance (NMR) spectra (Supplementary Figs. 8b–c) collectively demonstrate that the treated QDs possess a markedly lower and more homogeneous surface ligand density than the pristine and control ones. These results provide direct evidences for the displacement of oleylamine (OAm) ligands from the A-site by H₂S treatment. We therefore conclude that the H₂S not only occupies the X-site to form robust Pb–S bonds but also displaces the long-chain surface ligands at the A-site. This dual-site passivation strategy enables complete surface sulfidation, thereby reinforcing the structural integrity of CsPbI₃ QDs while simultaneously enhancing charge carrier transport.

We then performed theoretical calculations to elucidate the passivation mechanism of H₂S (Fig. 1e). The pristine CsPbI₃ QDs, initially capped by OAm ligands with an adsorption energy (E_{ads}) of –2.43 eV, expose abundant surface defects such as iodine and cesium vacancies (V_{I} or V_{Cs}). Our calculations show that H₂S preferentially fills these vacancies, particularly V_{I} ($E_{\text{ads}} = -2.98$ eV), forming a Pb–S bond (Supplementary Figs. 9 and 10). Notably, H₂S adsorption triggers the desorption of adjacent OAm ($E_{\text{des}} = 1.84$ eV), which in turn generates cesium vacancies (V_{Cs}). These newly created A-site vacancies are then filled by additional H₂S molecules, ultimately forming a dimeric structure (H₃S⁺–HS[–]) that passivates both A- and X-sites and yielding substantially reduced defect density. Adsorption in the form of (H₂S)₂ dimer shows markedly superior E_{ads} (–3.55 eV) over isolated H₂S molecular adsorption. This cooperative binding mode collectively contributes to a robust CsPbI₃ QD surface with enhanced structural integrity (Supplementary Note 1).

We further probed the optical and structural characteristics of CsPbI₃ QD films to evaluate the H₂S passivation effect. Grazing-incidence wide-angle X-ray scattering (GIWAXS) patterns (Supplementary Fig. 11) demonstrate superior structural order in the treated QD film, which exhibits the most continuous superlattice with minimal discrete scattering²⁷. This enhanced structural order is critical for achieving high-performance optoelectronic devices^{1,28}. We further evaluated the moisture stability of these QD films through contact angle measurements (Supplementary Fig. 12). The pristine and control films exhibit larger initial water contact angles than the treated film,

consistent with the presence of hydrophobic long-chain organic ligands that are attenuated after 12 hours H₂S treatment. Notably, the treated film demonstrates superior contact angle retention over time, indicating enhanced water resistance. This behavior is ascribed to the complete surface sulfidation, which improves its structural integrity and slows moisture interaction kinetics. Furthermore, temperature-dependent PL profiles (Supplementary Fig. 13) show the treated film possesses distinctly improved thermal stability, retaining higher initial intensity and slower emission decay.

The complete surface sulfidation not only reinforces the structural integrity of CsPbI₃ QDs but also plays a critical role in mitigating ion migration. We quantified this effect by measuring the ion migration activation energy (E_a) in QD films using temperature- and time-dependent current decay measurements (Supplementary Fig. 14 and Supplementary Note 2)^{29,30}. Significantly, the treated devices exhibit a substantially higher E_a of 374 meV, compared to 265 and 273 meV for the pristine and control ones, respectively (Figs. 2a–c). This elevated barrier reveals that exclusive X-site passivation is insufficient to address the lattice instability stemming from A-site ligand desorption, a process that also drives severe ion migration. Therefore, effective stabilization necessitates complete surface sulfidation of both sites, which concurrently enhances lattice robustness and minimizes ion migration. To directly observe ion migration, we subjected lateral devices to an electric field of 2.0 V·μm⁻¹ for 4 hours (Figs. 2d–f). Microscopic analysis reveals pronounced structural degradation in the pristine and control devices under applied voltage, characterized by widespread non-emissive pinholes across the film and severe fluorescence quenching. In contrast, the treated device maintains excellent structural integrity and luminescent homogeneity, demonstrating effective inhibition of ion migration.

The typically smaller effective electron mass (m_e^*) than effective hole mass (m_h^*) in perovskite QDs lowers the electron injection barrier in LEDs³¹. This imbalanced charge injection, thereby, fosters electron accumulation within the QDs, and subsequent nonradiative Auger recombination process involving two electrons and one hole. In pristine QDs, electron-only devices exhibit significantly higher current density than hole-only devices throughout the voltage range (Fig. 2g), confirming a substantially

lower electron injection barrier. This is further corroborated by an order-of-magnitude higher electron mobility than hole mobility (Supplementary Figs. 15a, b). Such imbalanced carrier injection thus drives severe Auger recombination and efficiency roll-off, resulting in a pronounced reduction in EQE under high current and brightness. However, this issue remains largely unaddressed in the control device (Fig. 2h, Supplementary Figs. 15c, d). In contrast, treated QDs enable markedly balanced charge injection, as evidenced by the minimal difference in current density between electron- and hole-only devices in the 4–8 V range (Fig. 2i). This barrier alignment is key to mitigating Auger recombination and roll-off, a conclusion further supported by the comparable electron and hole mobilities in these QDs (Supplementary Figs. 15e, f). To elucidate the origin of this improvement, we performed theoretical calculations (Supplementary Fig. 16). The treated QDs exhibit a slight increase in m_e^* and a marked reduction in m_h^* , accounting for the reduced hole injection barrier. The resulting reduced effective mass (m^*) is consequently smaller, which lowers the E_b of treated QDs⁴. Temperature-dependent PL spectroscopy further confirms the substantially reduced E_b in the treated QDs (74.6 meV) compared to the pristine (115.4 meV) and control (178.3 meV) ones, as shown in Supplementary Fig. 17. The lowest E_b in treated QDs weakens the Coulombic electron–hole attraction, thereby promoting more homogeneous carrier spatial distribution and thus suppressing Auger recombination³².

Carrier recombination dynamics

Motivated by the superior properties of treated QDs, we probed their carrier recombination dynamics. Generally, the reduction in E_b , while beneficial for suppressing Auger recombination, comes at the cost of reduced first-order exciton recombination at low excitation, which can be detrimental to LED performance³³. Therefore, suppressing trap-assisted recombination rate is essential to mitigate the performance limitations imposed by reduced E_b . We then characterized recombination dynamics under low excitation, the theoretical PLQY(N) and PL lifetime (τ) can be given by the well-known equations (Supplementary Note 3)^{16,34,35}:

$$\text{PLQY}(N) = \frac{k_{\text{exction}}}{k_{\text{exction}} + k_{\text{trap}}} \quad (1)$$

$$\frac{1}{\tau} = k_{exciton} + k_{trap} \quad (2)$$

Where $k_{exciton}$ represents the monomolecular radiative exciton recombination constant, k_{trap} is the first-order trap-assistant recombination constant. The PLQY at low excitation intensities is governed by the competition between radiative exciton recombination and trap-assisted nonradiative recombination. As shown in Figs. 3a–c, surface sulfidation dramatically reshapes the recombination kinetics. Compared to pristine QDs ($k_{exciton} = 4.12 \times 10^6 \text{ s}^{-1}$; $k_{trap} = 5.46 \times 10^6 \text{ s}^{-1}$), control QDs with only X-site sulfidation show a modest enhancement in $k_{exciton}$ ($5.86 \times 10^6 \text{ s}^{-1}$) and a dilution of k_{trap} ($4.24 \times 10^6 \text{ s}^{-1}$). In stark contrast, complete dual-site surface sulfidation (treated QDs) boosts $k_{exciton}$ by a factor of ~ 2.4 ($9.71 \times 10^6 \text{ s}^{-1}$) and suppresses k_{trap} by an order of magnitude ($5.11 \times 10^5 \text{ s}^{-1}$). These results demonstrate that the complete dual-site surface sulfidation treatment enables a reduction in E_b along with a substantial decrease in surface defect density, thus favoring radiative recombination pathways in the treated QDs.

At elevated excitation fluences, accelerated decay components emerge in the PL dynamics and gradually dominate, leading to a reduction in the effective lifetime (Figs. 3d–f). These fast decays correspond to a combination of bimolecular radiative recombination and trimolecular nonradiative Auger recombination processes³⁶. Multi-exponential fitting reveals a significant evolution in the Auger recombination proportion (fastest component, τ_3), which increases slightly from 54% (pristine) to 58% (control) but then drops markedly to 28% in the treated QDs. To further corroborate the changes in Auger recombination dynamics, we employed femtosecond transient absorption (TA) spectroscopy across a range of excitation fluences (Supplementary Fig. 18)³⁴. We obtained the biexciton recombination kinetics by subtracting the normalized traces at $1.4 \mu\text{W} \cdot \text{cm}^{-2}$ from the $12.3 \mu\text{W} \cdot \text{cm}^{-2}$ data (Figs. 3g–i). Single-exponential fitting of these kinetics yields biexciton lifetimes (τ_{xx}) of 293.78, 151.29, and 536.04 ps for pristine, control, and treated QDs, respectively. The Auger lifetime (τ_{Aug}) is determined using the relation $1/\tau_{xx} = 1/\tau_{xx,r} + 1/\tau_{Aug}$, where the radiative biexciton lifetime $\tau_{xx,r}$ is taken as one-fourth of the single exciton lifetime³⁷. Based on this relation, we obtained τ_{Aug} values of 297.13, 152.22, and 548.06 ps for pristine, control, and treated

QDs, respectively. Interestingly, while Auger recombination is aggravated in the control QDs compared to the pristine ones, it is strongly suppressed in the treated QDs³⁸. This trend, consistent with the data in Figs. 3d–f, underscores that complete dual-site surface sulfidation is pivotal for suppressing Auger recombination, whereas X-site-only modification is insufficient.

We further quantified the Auger recombination rate through TA measurements. The measured recombination rate dn/dt is governed exclusively by the instantaneous excitation density $n(t)$ ³⁹. The ground-state bleach region of the TA spectra is spectrally integrated to obtain a signal proportional to the excitation density $n(t)$ (Supplementary Fig. 19). Furthermore, the linear dependence of the initial TA amplitude on pump power confirms this signal serves as a reliable proxy for the carrier density. The TA kinetics is scaled by the initial carrier density (n_0 , see Supplementary Note 4 for estimation) to obtain the time-dependent carrier density $n(t)$. The carrier density-dependent recombination rates for various QDs are then determined by plotting the time derivative dn/dt against $n(t)$ (Figs. 3j–l). Monomolecular (*a*), bimolecular (*b*), and Auger (*c*) recombination are governed by recombination rates that scale linearly, quadratically, and cubically with carrier density, respectively. We observe a sequential increase in the *a* values (3.68 , 3.97 , and $4.11 \times 10^7 \text{ s}^{-1}$ for pristine, control and treated QDs, respectively), which is attributed to reduced trap density. This result aligns with the low-fluence transient PL kinetics shown in Figs. 3a–c. Moreover, the treated QDs demonstrate enhanced dominance of bimolecular radiative recombination across a broader carrier density range (1×10^{15} to $4 \times 10^{16} \text{ cm}^{-3}$), enabling sustained high PLQY. Notably, the treated QDs exhibit a significantly reduced Auger recombination rate ($1.09 \times 10^{-28} \text{ cm}^6 \text{ s}^{-1}$, compared to 3.18 and $3.37 \times 10^{-28} \text{ cm}^6 \text{ s}^{-1}$ for pristine and control ones, respectively) and a higher threshold carrier density ($\sim 4 \times 10^{16} \text{ cm}^{-3}$) for the onset of Auger-dominated recombination. These results collectively demonstrate the marked suppression of Auger recombination with complete dual-site surface sulfidation.

High-performance LEDs with ultralow roll-off

We fabricated LEDs using the CsPbI₃ QDs as emissive layer with the structure

shown in Supplementary Fig. 20a. The flat-band energy diagrams, constructed from ultraviolet photoelectron spectroscopy (UPS) and optical measurements (Supplementary Figs. 20b–h), reveal an advantageous upshift in the band level of the treated QDs, coupled with a reduction in its n-type doping. These modifications synergistically facilitate improved hole injection in the corresponding electroluminescent devices⁴⁰. Fig. 4a presents the current density–voltage and luminance–voltage characteristics of an optimally performing device following complete dual-site surface sulfidation (treated). This treatment enhances charge transport capability, increasing the maximum luminance ($\text{luminance}_{\text{max}}$) from 866 cd m^{-2} (pristine) to 7,779 cd m^{-2} , as illustrated in Supplementary Fig. 21. Significantly, the treated device demonstrates remarkably suppressed efficiency roll-off at high current density. As shown in Fig. 4b, the device reaches a maximum EQE of 30.71% at 52 mA cm^{-2} and retains high values of 29.14%, 24.04%, and 18.59% at elevated current densities of 104, 295, and 501 mA cm^{-2} , respectively, demonstrating minimal efficiency roll-off at high current density. For comparison, although the control QD-based device achieves a significantly higher peak EQE (21.09%) than the pristine device (6.11%), both exhibit similarly severe efficiency roll-off (Supplementary Fig. 22). These observations further highlight complete dual-site sulfidation as essential for suppressing Auger recombination, whereas X-site-only passivation proves ineffective.

EQE versus luminance curve for the treated device is illustrated in Fig. 4c, which shows the EQE_{max} at high brightness of 1,089 cd m^{-2} , and preserves a high EQE of 16.36% at its $\text{luminance}_{\text{max}}$ of 7,779 cd m^{-2} . Furthermore, a high EQE of ~30% is maintained across a luminance range of 100 to 2,000 cd m^{-2} . This exceptional ability to maintain high efficiency under high brightness makes this device particularly suitable for augmented and virtual reality (AR/VR), Micro-LED, and outdoor displays where both high luminance and energy efficiency are critical⁴¹. The treated device exhibits substantially enhanced operational stability, with operational half-lifetimes (T_{50}) of 1,331 min and 414 min at initial luminances of 113 cd m^{-2} and 990 cd m^{-2} (Fig. 4d, Supplementary Fig. 23), respectively—representing a nearly 50-fold improvement over the control device (27 minutes at 99 cd m^{-2}).

We statistically evaluated the distribution of EQE across a set of ten LEDs fabricated from treated QDs under high current density, as shown in Fig. 4e and Supplementary Table 1. These devices consistently maintain high EQE under high current densities, with average EQE values of 26.18%, 25.59%, and 19.70% measured at approximately 100, 300, and 500 mA cm⁻², respectively. We further analyzed the luminance at EQE_{max} and EQE at luminance_{max} of these devices. As illustrated in Fig. 4f and Supplementary Table 2, the EQE_{max} values are achieved at high luminance for all of these devices, with average and peak luminance of 3,441 and 5,891 cd m⁻², respectively. Moreover, these devices also maintain high EQE at luminance_{max}, with average and peak EQE of 16.23% and 19.27%, respectively. These results demonstrate preeminent reproducibility of our approach in fabricating LEDs with high performance and significantly suppressed efficiency roll-off. We benchmarked our device against leading red-emitting perovskite LEDs, with direct comparisons shown in Figs. 4g–i and summarized in Supplementary Table 3-5^{2,3,12,42-49}. In short, our devices exhibit significantly suppressed efficiency roll-off, maintaining exceptional EQE under both high current density and high luminance conditions.

Conclusion

We achieve an exceptionally robust surface on CsPbI₃ QDs through complete dual-site surface sulfidation. This process, mediated by (H₂S)₂ dimer, proceeds under mild conditions without damaging the QD structure. The resulting QDs exhibit suppressed Auger recombination, increased ion migration barriers, and balanced charge injection. LEDs devices fabricated from these QDs achieve a high EQE of 30.71%, along with significantly mitigated efficiency roll-off, maintaining high EQE under both high current density and high brightness (maintaining EQEs of 18.59% at ~500 mA cm⁻² and 25.01% at ~5,000 cd m⁻²). These devices demonstrate strong competitiveness and practical applicability. This work establishes (H₂S)₂-dimer-mediated dual-site surface sulfidation as a promising strategy for developing next-generation passivation approaches, opening pathways toward durable and high-performance perovskite optoelectronics.

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Methods

Materials

Lead iodide (PbI_2 , 99.999%, Sigma-Aldrich), Cesium carbonate (Cs_2CO_3 , 99.99%, Aladdin), oleylamine (OAm, 70-80%, Aladdin), oleic acid (OA, 90%, Aladdin), 1-octadecene (ODE, 90%, Sigma Aldrich), methyl acetate (MeAc, anhydrous grade, 99.5%, Aladdin), chlorobenzene (99.8%, Aladdin), dimethyl sulfoxide (DMSO, 99.9%, Aladdin), lithium fluoride (LiF , 99.5%, Aladdin). 1,3,5-Tris(1-phenyl-1H-benzimidazol-2-yl) benzene (TPBi), Poly (ethylenedioxy thiophene): polystyrenesulfonate (PEDOT: PSS, Heraeus-Clevios PVP AI4083), and Poly (N,N'-bis(4-butylphenyl)-N,N'-bis(phenyl)-benzidine) (Poly-TPD) were purchased from Xi'an Yuri Solar Co, Ltd. Hydrogen sulfide (H_2S , 5 ppm $\text{H}_2\text{S}/\text{N}_2$ gas mixture). All chemicals were used as received without further purification.

Preparation of Cs-OA precursor

Cesium oleate was prepared by adding Cs_2CO_3 (2.5 mmol), OA (5.0 mL), and ODE (40.0 mL) into a 100 mL three-neck flask. The mixture was degassed and dried under vacuum for 1 hour at 120 °C, then heated to 150 °C under N_2 until a clear solution was obtained.

Preparation of CsPbI₃ QDs

461.0 mg PbI_2 , 20.0 mL ODE, and 13.9 mg MgI_2 were loaded into a 50 mL three-neck flask, the mixture was then degassed and dried by applying vacuum for 60 min at 120 °C. 2.0 mL OAm and 2.0 mL OA were quickly injected, the temperature was raised to 170 °C, and 2.0 mL Cs-OA solution was quickly injected. After 5 s, the reaction solution was cooled to room temperature in an ice-water bath. The reaction product was separated by centrifugation for 5 min at 12000 rpm. The precipitate was dispersed in 1.0 mL mixed solvent of DMSO and toluene (1:800), and then 3.0 mL MeAc was added, the mixture was centrifuged for 5 min at 12000 rpm, the precipitate was redispersed in 1.0 mL octane. The 2.0 mL MeAc was added to the above solution, the mixture was centrifuged for 5 min at 12000 rpm. The precipitate was redispersed in 1.0 mL octane and centrifuged for 2 min at 5000 rpm. Let it stand for 12 hours before using it in light-

emitting devices (LEDs).

H₂S treatment

The purified QD solution (0.5 mL) was placed in a sealed vial and purged with N₂, 1.0 mL of H₂S was injected into the vial. Then, the solution was oscillated for 1 min and kept in dark at 4 °C for 12 hours to complete the sulfidation.

Fabrication of LEDs devices

Patterned ITO-coated glasses were cleaned successively using soap, deionized water, isopropanol, and acetone. Then ITO glasses were subjected to UV-ozone treatment for 20 min. PEDOT:PSS was spin coated onto the ITO substrate at 4000 rpm for 40 seconds and annealed in air at 150 °C for 20 min. The substrate was transferred into an N₂-filled glove box. Poly-TPD (5-15 mg/ml) was spin-coated onto the PEDOT:PSS film at 4000 rpm and annealed at 150 °C for 20 min. The perovskite QDs were spin-coated at 2000 rpm for 40 s. TPBi, LiF, and Al layers were sequentially deposited by thermal evaporation in a vacuum deposition chamber ($\sim 1 \times 10^{-4}$ Pa).

Electroluminescence characterization of LEDs devices

The active area of LED device was 4 mm². The electroluminescence was tested with a source meter (Keithley 2400), an integrating sphere (50 mm in diameter; Shenzhen Pynect) and a spectrometer (QE Pro, Ocean Optics) in a nitrogen-filled glovebox at room temperature. The absolute electroluminescence power spectra were collected using an integrating sphere and an Ocean Optics USB4000 spectrometer by mounting the devices on the wall of the integrating sphere. The EQEs were then calculated through the measured absolute electroluminescence power spectra and the current density. The devices were mounted on the open aperture of the integrating sphere to allow the light emitted from the glass surface to be collected, whereas the emission from the substrate edges was not collected. We have reproduced the measurements several times and also for several repetitions of the same experiment. LEDs were driven using a Keithley 2400 source meter at a constant current, and the luminance intensity was measured with a QE Pro calibrated spectrometer (Ocean Optics). The current used to drive the LED for stability tests was first determined using an EQE measurement system. The radiance of the spectrometer (QE pro, Ocean Optics) used in our system

was calibrated by a radiometric-calibrated light source (HL-3P-INT-CAL, Ocean Optics).

EXAFS, XPS, UPS, FTIR, and NMR measurements

EXAFS spectra were recorded at the BL13SSW beamline, Shanghai Synchrotron Radiation Facility (SSRF). The UPS and XPS spectra were obtained using a spectrometer (ESCALAB Xi+, Thermo Scientific) with an Al $K\alpha$ radiation source (1,486.6 eV). A monochromatic Al $K\alpha$ line (1,486.6 eV) was used for XPS, and He I radiation (21.2 eV) was used for ultraviolet photoelectron spectroscopy. The XPS and UPS spectra were recorded and analysed using the software Thermo Avantage. FTIR spectroscopic measurements were performed at the BL06B beamline, SSRF. NMR spectra were collected using Bruker AVANCE III (400MHz). To account for aging effects, all QD samples underwent a consistent 12-hour post-synthesis period prior to characterization or treatment. Specifically, the pristine sample was characterized after this 12-hour period; the control sample received its 1-minute sulfidation treatment at the 12-hour mark and was then immediately characterized; and the treated sample underwent the full 12-hour sulfidation process.

PL, PL lifetime, and UV-vis absorption spectra measurements

PL and PL lifetimes were obtained by using an EI-FLS1000 fluorescence spectrometer (Edinburgh Instruments) equipped with an integrating sphere. The time-resolved photoluminescence measurements were carried out using a nanosecond flash lamp as an excitation source ($\lambda_{\text{ex}} = 375$ nm). Steady-state absolute PLQYs were measured using a spectrofluorometer comprising a mountable integrating sphere and a 150 W xenon lamp. A temperature variable platform (Linkam THMS600) and liquid nitrogen were used for the temperature-dependent photoluminescence measurements. UV-vis absorption spectra were obtained using a Specord 200 Plus spectrophotometer.

TEM and XRD measurements

TEM observations were conducted using a Hitachi HT-7700 microscope operated at 100 kV. HRTEM observations and high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) images on the QDs were conducted using a JEMF2000 microscope operated at 200 kV. The TEM and HRTEM samples were

prepared by dropping the suspension onto carbon-coated copper grids. X-ray diffraction patterns were collected on a diffractometer (Bruker D8 Advance) with Cu K α radiation as the X-ray source, which ranged from 5° to 50° at a scanning rate of 3° min⁻¹.

GIWAXS measurement

The in situ GIWAXS measurements were performed at the BL17B1 beamline, SSRF. The monochromatic energy of the X-ray source was 18 keV, and the incidence angle was 0.2°. The sample-to-detector distance was calibrated with a lanthanum hexaboride (LaB₆) sample.

Transient absorption measurement

The output of a Ti: sapphire amplifier system (Spectra Physics Solstice Ace) operating at 1 kHz and generating approximately 100 fs pulses was split into the pump and probe beam paths. The 400 nm pump pulses were created by sending the 800 nm fundamental beam from the amplifier system through a second-harmonic-generating beta barium borate crystal of thickness 1 mm (Eksma Optics). Wavelength-tunable pump pulses (for example, 630 nm pump) were created by sending the 800 nm fundamental beam through an optical parametric amplifier (TOPAS). The pump was blocked by a chopper wheel rotating at 500 Hz while a computer operated a mechanical delay stage (Newport XPS-C8) to adjust the delay between the pump and the probe. The visible broadband beam (520–780 nm) was generated in a home-built noncollinear optical parametric amplifier, and the white light was split into two identical beams (probe and reference) by a 50/50 beam splitter. The reference beam, which passed through the sample, did not interact with the pump, allowing us to correct for any shot-to-shot fluctuations in the probe that would otherwise greatly increase the structured noise in the experiments. Based on this arrangement, small signals with a $\Delta T/T \approx 10^{-5}$ could be measured. The transmitted probe and reference pulses were collected with an InGaAs dual-line array detector (Hamamatsu G11608-512DA and Andor Shamrock SR-303i-B spectrograph) and driven and read out by a custom-built board (Stresing Entwicklungsbüro).

DFT calculations

DFT calculations were performed with the Device Studio (DS-PAW) from HZWTECH. The exchange-correlation energy was described by the Perdew–Burke–Ernzerhof form

of the generalized-gradient approximation exchange-correlation energy functional. The structural optimizations of the perovskite bulk and corresponding (1 0 0) surface with or without I vacancy defects were carried out by allowing all atomic positions to vary and fixing the lattice parameters until the energy difference of successive atom configurations was less than 10^{-6} eV. To ensure the calculation accuracy, the $3 \times 3 \times 3$ supercell was selected to constitute the bulk and surface structures. The force on each atom in the relaxed structures was less than $0.015 \text{ eV } \text{\AA}^{-1}$. The cutoff energy for the plane-wave basis set was set to 500 eV. The k-point spacing was set to be smaller than $0.03 \text{ \AA}^{-1}$ over the Brillouin zone. The electrostatic potential distribution of H₂S dimers was calculated under gas-phase conditions using Materials Studio (MS) based on density functional theory (DFT). Geometric optimization and single-point energy calculations employed PBE functionals combined with Grimme D3 dispersion correction (PBE-D3). A double-zeta polarization (DZP) basis set was used to ensure the reliability of charge distribution and interaction energies. The self-consistent field (SCF) density convergence threshold was set to 1×10^{-6} , and a fine-level integral mesh was used to improve potential surface smoothness. All calculations were performed with the solvent model disabled (gas-phase conditions).

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Author contributions: K.C conceived the idea. W.Z. initialized the project and contributed to the fabrication and characterization of QDs and devices. W.Z. and Y.Z. contributed to DFT calculations. W.Z., M.L., X.H., T.W., and X.Z. contributed to device fabrication and characterization. Optical properties were performed by W.Z. and Y.W. F.X., K.C., and G.L. directed and supervised the project. K.C. wrote the manuscript with input from all authors. All authors contributed to discussions and finalizing the manuscript.

Competing interests: The authors declare that they have no competing interests.

Data and materials availability: All data needed to evaluate the conclusions in the paper are present in the paper or the supplementary materials.

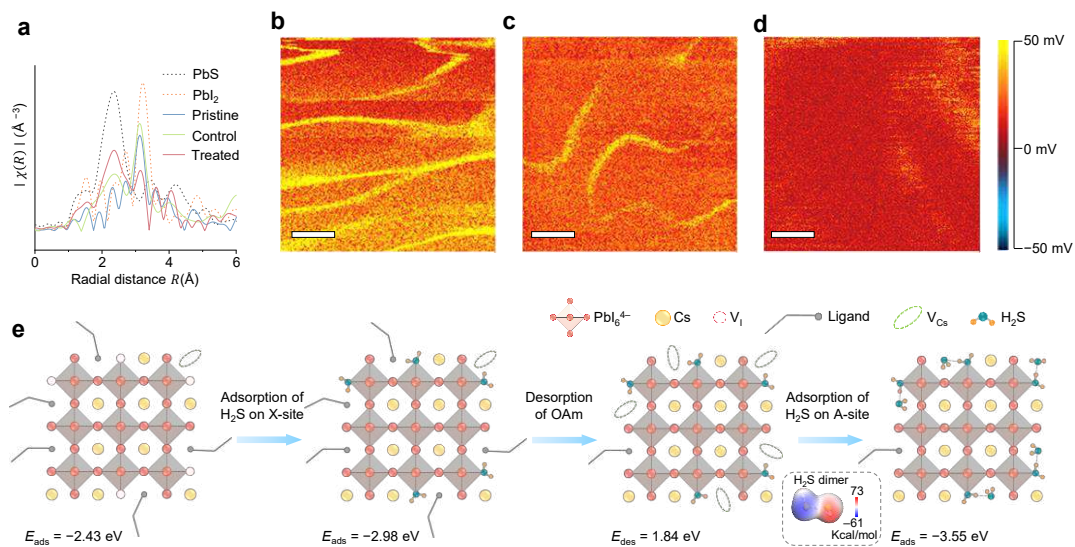


Fig. 1 | Dual-site surface passivation. (a) Pb L_3 -edge EXAFS spectra (R-space plots). (b to d) FTIR mapping of (b) pristine, (c) control, and (d) treated QDs. (e) Simplified schematic diagram showing the dual-site surface sulfidation modification process of CsPbI_3 QDs, inset: electrostatic potential mapping of $(\text{H}_2\text{S})_2$ dimer. Scale bars, 2 μm (b to d).

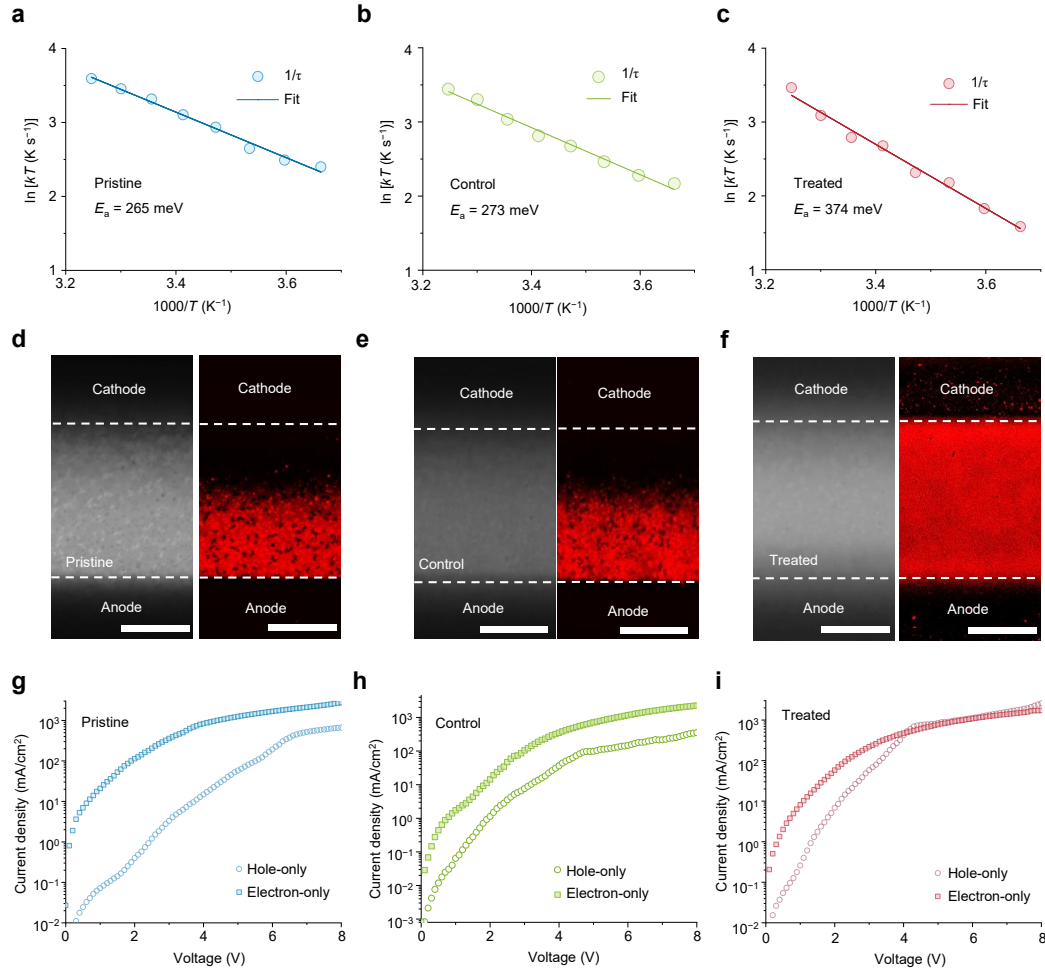


Fig. 2 | Optoelectronic properties. (a to c) Temperature-dependence plots of $\ln(kT)$ versus $1,000/T$ of (a) pristine, (b) control, and (c) treated QD films. (d to f) Fluorescence microscopy images of (d) pristine, (e) control, and (f) treated QD films after poling at $2.0 \text{ V } \mu\text{m}^{-1}$ for 4 h. (g to i) SCLC curves of (g) pristine, (h) control, and (i) treated QD films. Scale bars, $50 \text{ } \mu\text{m}$ (d to f).

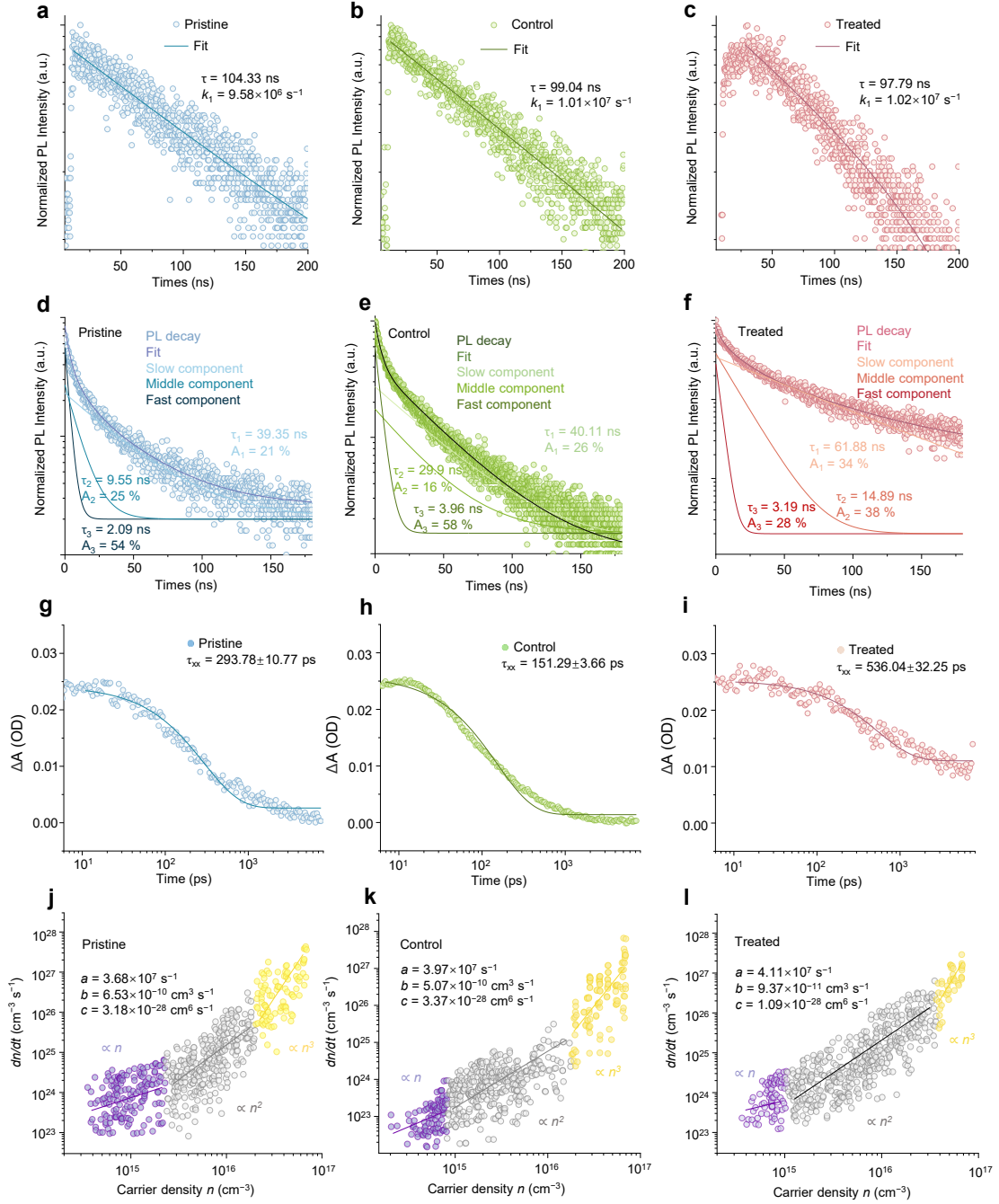


Fig. 3 | Carrier recombination dynamics. (a to c) TRPL decay curves of (a) pristine, (b) control, and (c) treated QDs under a low pump fluence of 2.94 nJ cm^{-2} . (d to f) TRPL decay curves under a high pump fluence of $7.23 \text{ }\mu\text{J cm}^{-2}$ for (d) pristine, (e) control, and (f) treated QDs. (g to i) Biexciton recombination TA kinetics traces of (g) pristine, (h) control, and (i) treated QD films obtained by subtraction between the kinetic traces normalized to the long-lived tail under 1.4 and $12.3 \text{ }\mu\text{W cm}^{-2}$ excitation fluences. (j to l) Recombination rate dn/dt over charge density n for of (j) pristine, (k) control, and (l) treated QD films. The solid lines are guides to the eye.

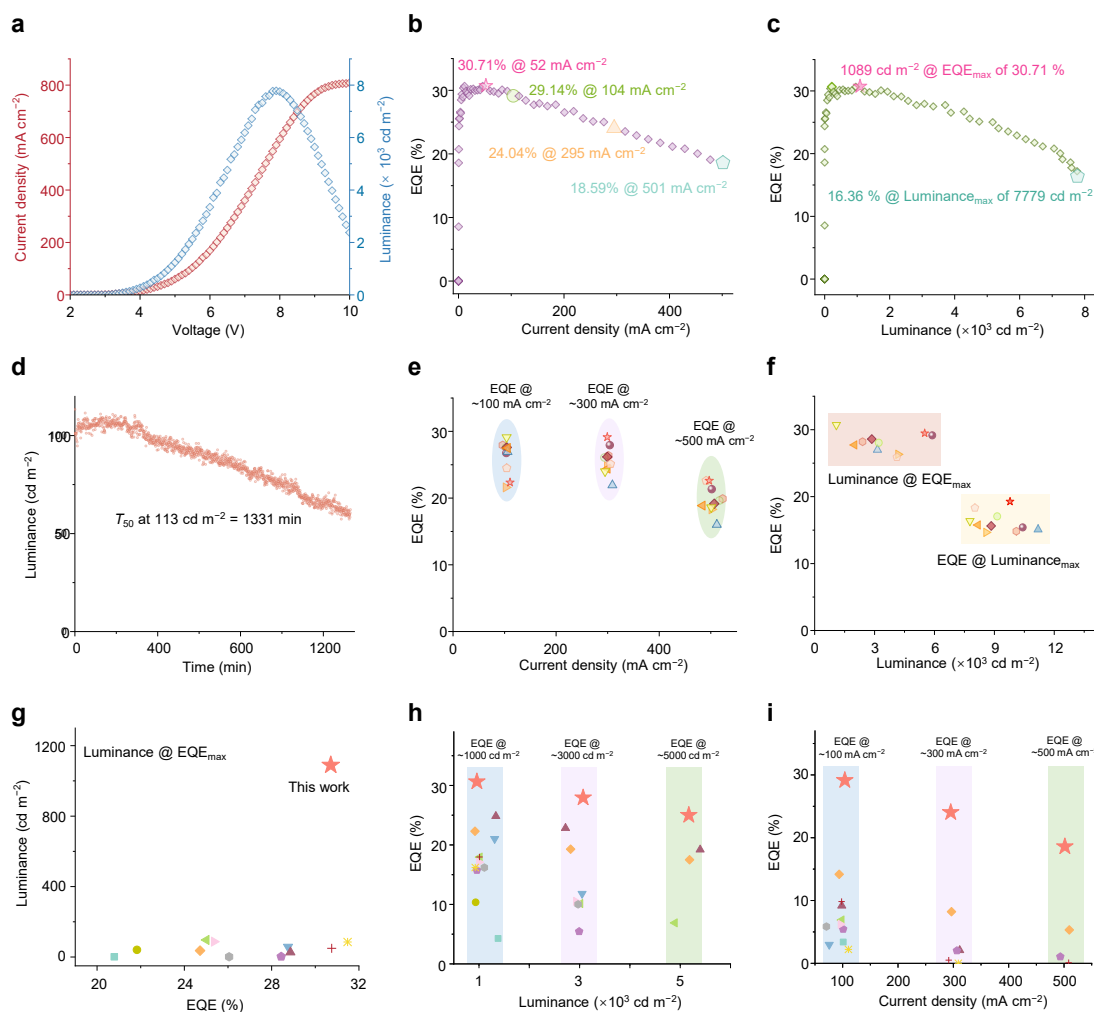


Fig. 4 | Device performance of treated CsPbI₃ QD-based LEDs. (a) Typical current density–voltage–luminance curve of the LED. (b) EQE–current density curve of the LED. (c) EQE–luminance curve of the LED. (d) T_{50} operational lifetime of the LED ($L_0 = 113 \text{ cd m}^{-2}$). (e) EQE versus various high current density collected from 10 LEDs. (f) luminance at EQE_{max} and EQE at luminance_{max} of corresponding 10 devices in panel (E). (g to i) Summaries of the (g) luminance at EQE_{max}, (h) EQE at high current densities, and (i) EQE at high luminance of the reported red-emitting perovskite LEDs and our LEDs (Supplementary Table 3–5).

Supplementary Files

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