

Over 30% efficient and highly stable Co-SnO₂-based QLEDs through ultrathin MgCl₂ interface engineering

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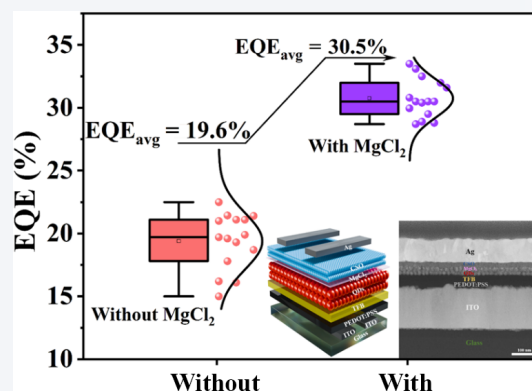
ABSTRACT: SnO₂-based quantum dot light-emitting diodes (QLEDs) offer superior environmental stability over ZnO-based devices, yet their electroluminescent performance is often limited by interfacial exciton quenching and unbalanced carrier injection. Here, we employed solution-processed Co-doped SnO₂ (CSO) nanoparticles as the electron transport layer to reduce oxygen vacancies and optimize energy-level alignment. As a result, the red QLED with 5 wt.% CSO achieved an external quantum efficiency (EQE) of 21.5% at a luminance of 187 cd/m². However, at high luminance the carrier-balance improvement cannot be maintained and the CSO-device still shows significant efficiency roll-off. To further optimize the interface, we introduced an ultrathin MgCl₂ interlayer between the quantum dot layer and the CSO layer. The MgCl₂ layer acts as a functional modifier, passivating surface defects, tuning energy levels, and improving carrier balance. Consequently, the device achieved a maximum EQE exceeding 30% and a long T₉₅ operational lifetime (at 1000 cd/m²) of 3367.7 h. This synergistic strategy provides a practical pathway toward high-performance and stable QLEDs.

KEYWORDS: external quantum efficiency (EQE), Co doped tin oxide, quantum dot light-emitting diodes (QLEDs), MgCl₂ interlayer, interface engineering

1 Introduction

Due to the superior color purity with emission linewidths as narrow as 20–30 nm, a broad color gamut exceeding 100% of the National Television Standards Committee (NTSC) standard, and high external quantum efficiency (EQE) of over 20%, quantum dot light-emitting diodes (QLEDs) have emerged as a revolutionary display technology, rapidly gaining prominence in both academic research and industrial applications [1–4]. A typical QLED is a sandwich multilayer structure with transport and emitting layers. The

electron transport layer (ETL) plays a dual role: facilitating efficient electron injection to optimize charge balance, while also serving as a protective barrier against oxygen, moisture, and ion migration, which are key factors in device degradation. Zinc oxide (ZnO) nanoparticles (NPs) have been widely used in QLEDs, owing to their high electron mobility (> 1 cm²/(V·s)) and suitable energy level [5, 6]. However, the high surface reactivity of ZnO NPs, driven by abundant oxygen vacancies and undercoordinated zinc sites, may inevitably induce photo-induced degradation. These may lead to the positive aging phenomenon [7]. To improve the shelf storage stability, the surface reactivity of ZnO has been modified through superficial treatments. Although strategies such as Al/Mg doping [8] and organic ligand passivation [9] have been employed, the inherent instability of ZnO remains a bottleneck for commercial applications. Alternative materials, including titanium dioxide (TiO₂) [10], tin dioxide (SnO₂) [11], and organic molecules [12] were studied. SnO₂ NPs stand out as a solution-processable n-type



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semiconductor with a compelling set of properties: a wide bandgap, high electron mobility, and excellent optical transparency [13, 14]. Furthermore, the stronger Sn=O bonds [15, 16] significantly suppress oxygen vacancy formation, thereby enhancing environmental stability. In addition, SnO₂-based ETLs can effectively alleviate the positive aging effect commonly observed in ZnO-based QLEDs, making SnO₂ a promising alternative electron transport material for achieving more stable device [17, 18].

Recent years have witnessed the development of SnO₂-based QLEDs. In 2020, Liu et al. proposed a quantum size engineering strategy by synthesizing size-reduced SnO₂ NPs, offering a promising ETL material for high-performance QLEDs [19]. In 2022, Chen et al. pioneered the use of tetramethylammonium hydroxide as a stabilizer, where its –OH groups form coordinated bonds with surface Sn atoms [17]. And QLEDs reached an EQE of 13.7%, along with a T_{95} lifetime of 3200 h (@1000 cd/m²). Recently, SnO₂-based ETLs have been widely used in QLEDs to eliminate the positive aging effects. In 2024, Gao et al. demonstrated an EQE of 12.9% red QLEDs with no positive aging by employing ligand-assisted SnO₂ ETLs [20]. However, electroluminescence (EL) performance of SnO₂-based QLEDs generally fell behind the mature ZnO-based QLEDs. One contributing factor is the presence of excessive hydroxyl groups introduced during SnO₂ synthesis, which can increase surface defects, particularly at the SnO₂/QD interface. These defects may directly quench excitons and reduce the luminance efficiency. Additionally, the high electron mobility of SnO₂ could result in excess electrons, which can induce significant leakage current, or electron accumulation [21], thereby compromising both efficiency and operational stability. To improve the performance of SnO₂-based QLEDs, strategies primarily focus on tailoring the energy levels and charging carrier mobility by elemental doping. Luo et al. balanced carrier transport in green QLEDs with Zn-doped SnO₂ NPs, achieving an EQE of 5.03% and a T_{50} lifetime of 9141 h [21]. Ma et al. reported a Mn-doping strategy to enhance the Mn–O bond energy and the EQE of QLED was 11.4% and the T_{95} lifetime extended to 1009.2 h [22]. Zhao et al. incorporated Zr into SnO₂ to improve the dispersion and upshift the energy levels, ultimately raising an EQE of 16.6% [23]. Xia et al. focused on low-concentration Sb doping, revealing its ability to transform the conduction mechanism from trap-limited to space-charge-limited [24]. And the QLED reached a remarkable EQE of 18.3% and a T_{95} lifetime of 4320 h. While, surfaces of SnO₂ ETLs frequently exhibit numerous dangling bonds, defect states, or high-energy surfaces [11, 20]. These interfacial defects could quench the excitons via non-radiative energy transfer process [25–27]. Concurrently, Liu et al. demonstrated a Zn-doped SnO₂/GaMoO₄ heterostructure, where 4.8 eV wide bandgap GaMoO₄ acted as an energy barrier to prevent electron leakage [28]. This synergistic design achieved a record EQE of 27.1%, and a T_{95} lifetime of 1828.3 h.

However, the EQE of SnO₂-based QLEDs remains inferior to that of mature ZnO-based QLEDs. By decreasing electron mobility through vacancy passivation and deep-level trapping [29–32], herein, cobalt (Co) doped SnO₂ NPs (CSO) were synthesized to suppress the exciton quenching at QDs/ETL interface. Unlike some previously reported Zn-doping strategies that may introduce additional oxygen-vacancy-related defects in SnO₂ [19], Co doping can effectively suppress oxygen vacancies while simultaneously regulating carrier transport. Additionally, the Co-doped strategy fabricates to obtain a smoother film and effectively tailored the

energy level. At a luminance of 187 cd/m², the CSO-based QLED exhibited a significantly enhanced EQE of 21.5%, which is comparable to that of the state-of-the-art SnO₂-based QLEDs. However, at high luminance the carrier-balance improvement cannot be maintained and the CSO-device still shows significant efficiency roll-off. To further mitigate exciton quenching and regulate the QD/ETL interface, an ultrathin MgCl₂ interlayer was introduced. This work emphasizes the synergistic interfacial regulation between CSO ETL and MgCl₂ interfacial layer, including local interfacial dipole formation, electron injection modulation, and suppressed non-radiative recombination. The deployment of MgCl₂ as an interlayer capitalizes on its interface modification and defect passivation. Specifically, its Cl[–] ions passivate defects by saturating dangling bonds to suppress charge recombination, while Mg²⁺ ions can cooperatively tune surface energy levels [33, 34]. As a result, QLED with MgCl₂ reaches a record EQE of 33.1% for red SnO₂-based QLEDs. Moreover, lifetime evaluations of the MgCl₂-based devices revealed a T_{95} lifetime of 3367.7 h@1000 cd/m². This synergy of chemical and physical properties ultimately leads to remarkable enhancements in both the EL performance and operational stability, contributes to advancing SnO₂-based QLED technology toward commercialization.

2 Experimental section

2.1 Materials

Chlorobenzene (99.8%) and octane (99%) were purchased from Sigma-Aldrich. Tin(IV) chloride pentahydrate (SnCl₄·5H₂O, 99%), ethylene glycol (EG, 98%), and tetramethylammonium hydroxide pentahydrate (TMAH, 97%) were acquired from Aladdin. Cobalt acetate (Co(Ac)₂, 98%), acetic acid (HAc, 36%), ethyl acetate (99.5%), dimethyl sulfoxide (DMSO), and ethanol (99.8%) were purchased from Macklin. Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS, 4083) and poly[(9,9-dioctylfluorenyl-2,7-diyl)-alt-(4,4'-(N-(4-butylphenyl)diphenylamine)] (TFB, $M_w \approx 20,000$ –40,000) were purchased from Xi'an Polymer Light Technology. Red CdSe/ZnSe/ZnS QDs were obtained from Xingshuo Nanotech Co., Ltd. All materials were used as received without further purification.

2.2 Synthesis of SnO₂ NPs

SnO₂ NPs were synthesized via a solution-based method. Specifically, 2 mmol of SnCl₄·5H₂O was dissolved in 20 mL DMSO in a 100 mL three-neck flask and stirred for 10 min to form a clear precursor solution. Subsequently, 2 mL of HAc was added dropwise into this solution, followed by the slow dropwise addition of 7.25 mL TMAH·5H₂O solution (2 mol/L, EG). The mixture was stirred for another 30 min, yielding a clear and transparent solution. The resulting solution was then subjected to a reaction in a dimethyl silicone oil bath at 150 °C for 1 h. After the reaction, the flask was rapidly transferred to an ice-water bath and cooled to room temperature. The obtained mixture was purified three times via centrifugation using ethanol and ethyl acetate. The purified NPs were dispersed in ethanol to form a dispersion with a concentration of 40 mg/mL. To further enhance the dispersion stability, a TMAH solution in ethanol was added to this dispersion at a volume ratio of 1:4, with continuous stirring until a clear and well-dispersed SnO₂ NPs solution was obtained. For the synthesis of CSO NPs, the procedure was essentially identical to the steps above, with the only

modification of the precursor solution: $\text{Co}(\text{Ac})_2$ was added to 2.5% to 7.5% by weight of $\text{SnCl}_4 \cdot 5\text{H}_2\text{O}$.

2.3 Fabrication of QLEDs

The ITO substrates were cleaned, followed by spin-coating an PEDOT:PSS solution at 4000 rpm for 40 s in ambient air, and then annealed at 150 °C for 15 min. After transferring the substrates into a nitrogen-filled glovebox, the device was fabricated using layer-by-layer method, a TFB (8 mg/mL, chlorobenzene), CdSe QDs (20 mg/mL, octane), MgCl_2 (1 mg/mL, ethanol) and SnO_2 NPs (20 mg/mL, ethanol) were spin-coated at 3000, 2000, 4000, and 4000 rpm for 30, 30, 40 and 30 s, respectively. The TFB, CdSe QDs, MgCl_2 and SnO_2 NPs layer were annealed at 150, 90, 80, and 80 °C, respectively. Finally, a 100 nm Ag electrode was deposited via thermal evaporation under a high vacuum of 5×10^{-4} Pa. The device active area, defined by the overlap between the ITO anode and Ag cathode, was 4 mm².

2.4 Characterization

The X-ray diffraction (XRD) patterns were measured by a Bruker D8 Advance system with a $\text{Cu K}\alpha$ radiation ($\lambda = 1.54178 \text{ \AA}$) source. The X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS) measurements were carried out on Thermo Fisher Scientific (ESCALAB Xi+). The ultraviolet-visible (UV-vis) absorption spectra were obtained by a

UV-vis spectrophotometer (LAMBDA, 750 UV). Dynamic Light Scattering (DLS) measurements were conducted using nanoparticle size and zeta potential analyzer (HORIBA, SZ-100V2). The morphology and cross-section transmission electron microscope (TEM) images were obtained using the high-resolution mode by the TEM (JEOL, JEM F200). Atomic force microscopy (AFM) images were conducted using a scanning probe microscope (Shimadzu, SPM-9700). The photoluminescence (PL) and time-resolved PL (TRPL) spectra was analyzed using the Fluorolog[®]-3 and associated accessories. The thickness of the films was tested by a surface profiler (Alpha-Step, D-600). The IS spectra were tested using the AUTOLAB PGSTAT204 electrochemical workstation. The J - V - L and EQE- L curves, and lifetime were tested by an Xi Pu Optoelectronics QLED measurement system (XPQY-EQE) and lifetime measurement system (XP-LTS-8). EL spectra of the QLED devices were collected using a Keithley 2400 light source meter and an Ocean Optics QE65000 spectrometer.

3 Results and discussion

XRD patterns of different CSO are exhibited in Fig. 1(a), and the patterns confirmed the tetragonal rutile structure of SnO_2 (0 wt.% CSO, JCPDS card No. 41-1445). The diffraction peaks gradually broadened by increasing Co content, which can be attributed to the enhanced lattice strain [35]. The full-scan XPS is shown in Fig. S1

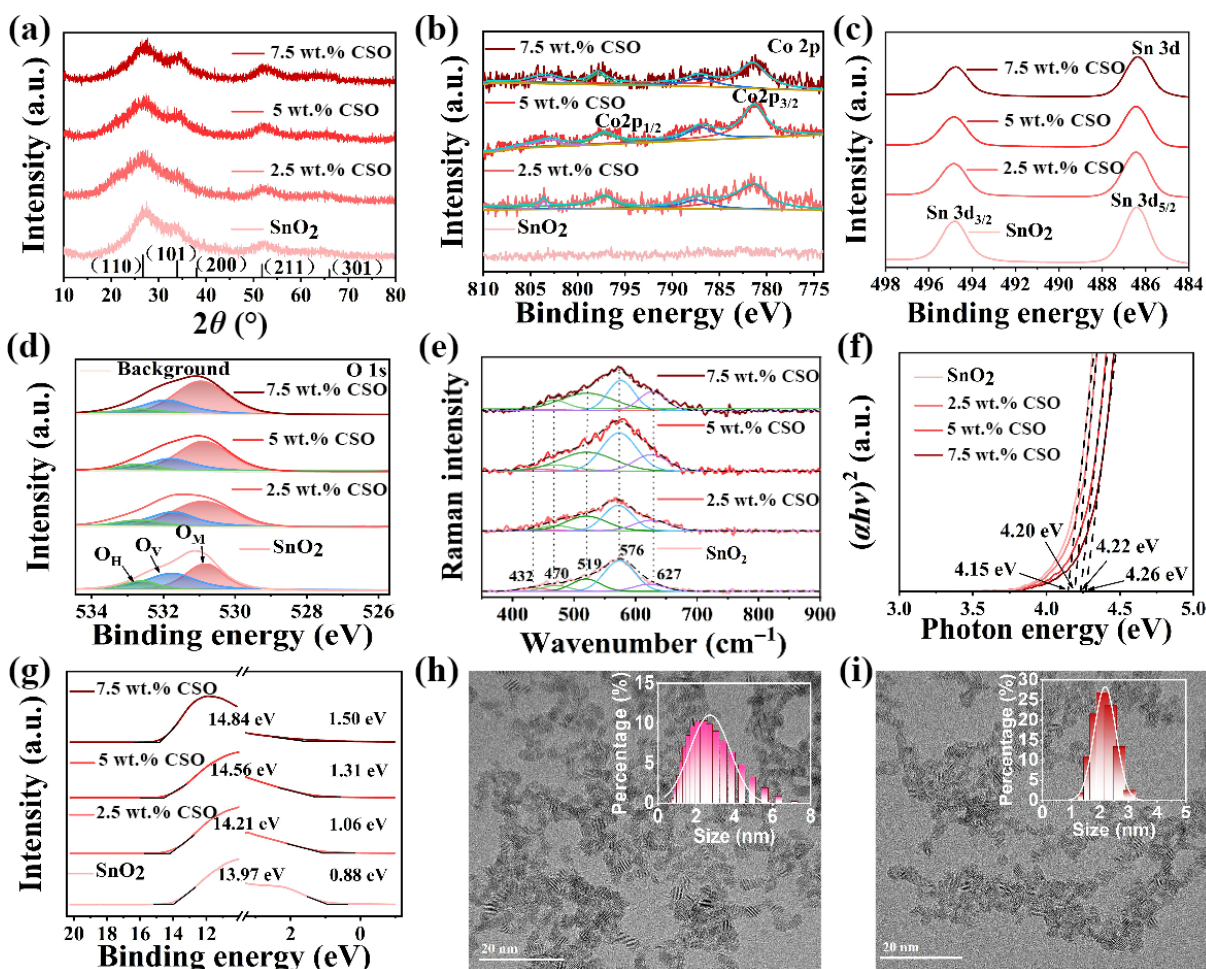


Figure 1 Characteristics of CSO with different Co concentration: (a) XRD patterns, high-resolution XPS spectra of (b) Co 2p, (c) Sn 3d, and (d) O 1s orbit, (e) Raman spectra, (f) Tauc plot, (g) UPS spectra. TEM images of (h) SnO_2 NPs and (i) 5.0 wt.% CSO, inset DLS spectra.

in the Electronic Supplementary Material (ESM)), and all revealed the presence of Sn and O. Furthermore, energy dispersive X-ray spectroscopy (EDS) characterization together with the corresponding statistical analysis (Fig. S2 and Table S1 in the ESM) demonstrates that the actual Co contents are in good agreement with the nominal doping concentrations (2.5 wt.%–7.5 wt.%), confirming the reliable incorporation of Co into the SnO₂ lattice. High-resolution XPS spectra of Co 2p orbit are provided in Fig. 1(b), two characteristic peaks located at 797.46 and 781.45 eV emerge in the 5 wt.% CSO, corresponding to Co 2p_{1/2} and Co 2p_{3/2}, respectively, indicating the presence of Co²⁺ in the NPs. The high-resolution Sn 3d orbit XPS spectra (Fig. 1(c)) of SnO₂ NPs exhibited two peaks at 486.36 and 494.85 eV, corresponding to Sn 3d_{5/2} and Sn 3d_{3/2}, respectively. By Co doping, the energy difference between Sn 3d_{5/2} and O 1s narrows by 0.2 eV, suggesting strong Sn-O-Co interactions. Additionally, the full width half maximum (FWHM) of the Sn 3d_{5/2} peak decreases from 1.55 to 1.1 eV (5.0 wt.% CSO), which can be attributed to the reduced proportion of Sn-O bonds. Detail information of the fitted Co 2p and Sn 3d characteristic peaks is collected in Table 1. The high-resolution XPS spectra of O 1s orbit are demonstrated in Fig. 1(d). The O 1s peak of CSO can be deconvoluted into three Gaussian distribution curves, corresponding to binding energies at approximately 530.81 eV (O_M, lattice oxygen), 531.77 eV (O_V, oxygen vacancy), and 532.68 eV (O_H, surface hydroxyl oxygen), respectively. The electrical conductivity of SnO₂ is closely related to the O_V concentration, as oxygen vacancies can act as shallow donor defects to provide free carriers, while excessive O_V may also introduce carrier scattering and defect-induced localization effects. As the relative ratio of O_V decreased (Table S2 in the ESM) and the broad green-yellow emission was notably quenched (Fig. S3 in the ESM), Co doping is suggested to suppress the O_V-related defects in SnO₂ [30]. Raman spectra (Fig. 1(e)) further support this conclusion, where the defect-related Raman feature gradually weakened with increasing Co concentration. The Raman mode located at approximately 574 cm⁻¹ can be assigned to the in-plane oxygen vacancy-related vibrational mode [30], whose intensity is generally correlated with the concentration of oxygen vacancies. Density functional theory calculations have suggested that this mode originates from in-plane oxygen vacancies in the surface region of SnO₂ nanoparticles [36]. In addition, the Raman peak at approximately 627 cm⁻¹ corresponds to the A_{1g} vibrational mode of rutile SnO₂, while the peak near 470 cm⁻¹ is assigned to the E_g mode. Previous studies have reported that the E_g mode exhibits a blue shift as the SnO₂ nanoparticle size decreases [30]. Beside these characteristic modes, additional Raman features located at approximately 430 and 540 cm⁻¹ are also observed. The peak around 540 cm⁻¹ is generally attributed to the Raman-forbidden B_{1u} mode activated by the relaxation of selection rules in nanostructured SnO₂ [37, 38]. Meanwhile, the new mode near 430 cm⁻¹ is associated with oxygen-vacancy clusters [39]. These additional modes are more

pronounced in smaller SnO₂ NPs and gradually weaken with increasing particle size.

Notably, the Raman mode associated with in-plane oxygen vacancies at 574 cm⁻¹ does not exhibit an obvious peak-position shift after Co doping, whereas the oxygen-vacancy-cluster-related mode at 430 cm⁻¹ shows a slight shift toward higher wavenumbers, indicating that Co doping modulates the local defect environment. More importantly, the intensities of both the 574 and 430 cm⁻¹ oxygen vacancy related modes gradually decrease with increasing Co doping concentration, suggesting that Co doping effectively suppresses oxygen vacancy related defects and vacancy clusters in SnO₂ films. This observation is consistent with the XPS analysis and further supports the defect suppression effect induced by Co doping. O_H is known to serve as exciton quenching centers, which may negatively impact the performance of QLED [21]. In contrast, the O_H content changes from 14.30% to 9.37% (5.0 wt.% CSO), which is beneficial for the efficient EL emission. It should be noted that oxygen vacancies in SnO₂ may play a dual role by simultaneously acting as electron donors and carrier scattering/trapping centers. In this work, the reduced electron current is mainly attributed to the decreased free carrier concentration caused by O_V suppression, together with the increased electron injection barrier after Co doping.

The Tauc plot (Fig. 1(f)) reveals a substantial widening of E_g with increasing Co concentration, which can be attributed to the quantum size effect. The significant blue-shift may be attributed to the formation of new electronic states within the intrinsic bandgap of SnO₂ [40]. The secondary electron cutoff (E_{cutoff}) in the UPS spectra (Fig. 1(g)) indicates a slight increase in the work function. And the valence band maximum (VBM) is determined according to our previous work [21]. The VBMs are 8.13, 8.07, 7.97, and 7.85 eV for 0 wt.%, 2.5 wt.%, 5.0 wt.%, and 7.5 wt.% CSO, respectively. The VBM shifts to the Fermi level, suggesting that Co doping influences the surface band bending and electron affinity. The conduction band maximums (CBMs) are summarized in Fig. S4 in the ESM. Co doping progressively shifts the VBM and CBM to higher energies. TEM images of CSO were shown in Figs. 1(h) and 1(i). The average size (D_{avg}) is determined by dynamic light scattering (DLS) spectra, and approximately 2.75 nm is obtained for SnO₂ NPs. With Co-doping, the D_{avg} decreased to 2.20 nm for 5.0 wt.% CSO. The uniform and smaller size NPs are expected to improved surface smoothness. AFM images are shown in Fig. S5 in the ESM. The root-mean-square roughness of 5.0 wt.% CSO film (0.807 nm) is notably lower than that of SnO₂ film (1.137 nm). This confirms that Co incorporation is beneficial for forming high-quality film, which is critical for minimizing leakage current and improving overall LED performance.

The optimized QLED employs a multilayer structure (Fig. 2(a)), in sequence: ITO served as anode, PEDOT:PSS as hole-injecting layer (HIL), TFB as hole transport layer (HTL), red CdSe QD as EML, CSO as ETL, and finally an Ag cathode. As observed in Fig. 2(b), the electron injection barrier increases with the use of CSO ETL. This elevated barrier effectively limits charge injection, thereby facilitating a more balanced carrier. The QDs exhibited a photoluminescence (PL) peak at 621 nm with a full width at half maximum (FWHM) of 23 nm and a photoluminescence quantum yield (PLQY) of 91.4% (Fig. S6 in the ESM). The normalized EL spectra (Fig. 2(c)) exhibit an emission peak at 630 nm with a FWHM of 26 nm. Compared with the PL peak of the pristine QDs, the EL emission showed an obvious redshift, which can be

Table 1 Fitted binding energies of Sn 3d and Co 2p orbitals for different CSO

Co concentrations (wt.%)	Sn 3d _{5/2} (eV)	Sn 3d _{3/2} (eV)	Co 2p _{1/2} (eV)	Co 2p _{3/2} (eV)
0	486.36	494.85	—	—
2.5	486.49	494.91	797.08	781.33
5	486.55	495.04	797.46	781.45
7.5	486.63	495.10	797.64	781.52

attributed to the quantum-confined Stark effect under an applied electric field [21]. As current density–voltage–luminance (J – V – L) characteristics shown in Fig. 2(d), the CSO-based QLEDs exhibit a lower current density than the control one at the same driving voltage, consistent with the suppressed electron injection and electron transport in the CSO ETL caused by the increased electron injection barrier and reduced carrier transport capability after Co doping (Fig. S7 and Table S3 in the ESM). The charge-regulation functionality of CSO is closely tied to the luminance level. In the low-to-moderate regime, a higher electron injection barrier curbs excessive electron flow, which enhances charge balance and boosts radiative recombination. At high driving voltages (6–7 V), the current densities of different CSO devices gradually become closer. This deliberate moderation of electron flow is pivotal, as it leads to a superior charge balance in the EML, culminating in a peak CE of 27.0 cd/A (5.0 wt.% CSO device, Fig. 2(e)). The maximum EQE for devices with 0 wt.%, 2.5 wt.%, 5.0 wt.%, and 7.5 wt.% CSO ETL are 9.4%, 15.1%, 21.5%, and 10.1%, respectively. The maximum EQE is mainly achieved at low luminance of 187 cd/m², while at high luminance/current density the carrier-balance improvement cannot be maintained and the CSO-device still shows significant efficiency roll-off.

Notably, the device performance is determined by the combined effects of oxygen-vacancy suppression, carrier transport capability, and charge balance. Although the 7.5 wt.% CSO exhibits the lowest oxygen vacancy concentration, excessive Co doping overly suppresses electron transport. In contrast, the 2.5 wt.% CSO retains relatively high electron mobility, but still cannot effectively suppress

excessive electron injection. Therefore, the 5 wt.% CSO achieves a more balanced regulation of defect passivation and electron transport, leading to improved charge balance and radiative recombination efficiency, as evidenced by its prolonged PL lifetime and highest EQE. However, under high driving voltages and high luminance conditions, the enhanced electric field weakens the limitation effect of the interfacial injection barrier, allowing excessive electrons to be injected again into the QD layer. Meanwhile, Auger non-radiative recombination and exciton quenching become more severe at high exciton densities, resulting in efficiency roll-off and the reduced EQE advantage of CSO-based devices at high luminance. EL performance of QLEDs with different ETL is summarized in Table S4 in the ESM. The effect of Co dopant was investigated by comparing the PL properties of glass/X wt.% CSO/QDs samples. TRPL measurements (Fig. 2(f)) reveal that the average PL lifetime (τ_{avg}) of QDs on the bare SnO₂ NP is only 11.2 ns, a substantial decrease from the 15.4 ns (QD film), primarily due to interfacial exciton quenching. However, as the Co concentrations increased to 5.0 wt.%, the τ_{avg} increased to 15.0 ns. This relative long lifetime was accompanied by higher PLQY and lower non-radiative decay rate, confirming that Co doping effectively passivates defects and suppresses exciton quenching. The details fitted τ_{avg} data is provided in Table S5 in the ESM. This suppression of non-radiative recombination also leads to a high steady-state PL intensity, as evidenced in Fig. 2(g). To evaluate the electron transport properties of the CSO NCs, electron-only device (EOD) was fabricated. As shown in Fig. 2(h), the current density of the CSO-based EODs is significantly lower than

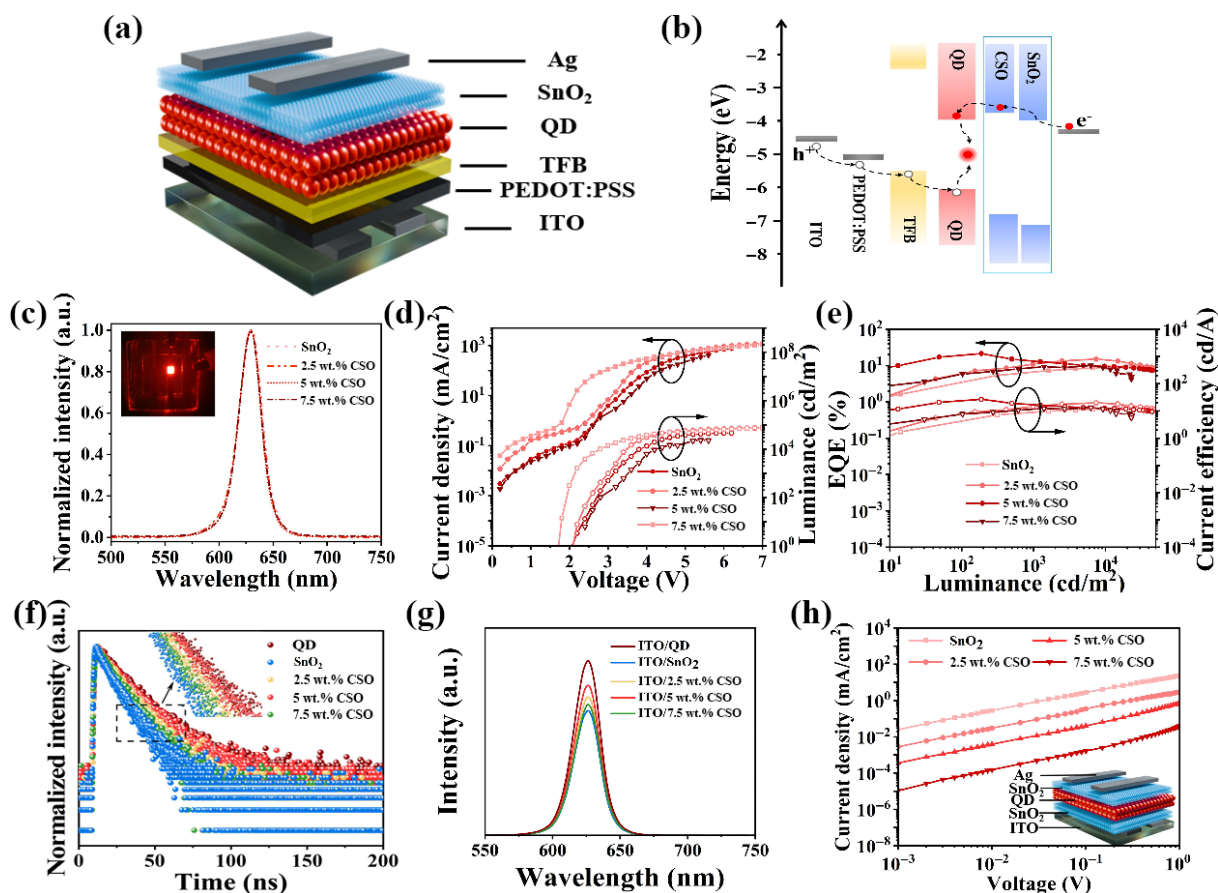


Figure 2 EL performance of QLED with different ETLs. (a) Structure architecture, (b) energy level diagram, (c) normalized EL spectra, (d) J – V – L curves, (e) EQE and CE as a function of luminance, (f) TRPL and (g) PL spectra of ITO/CSO/QDs samples, and (h) J – V curves of EODs.

that of the SnO_2 -based control. The use of an engineered CSO ETL with retarded electron transport serves to regulate electron injection, promoting optimal charge balance by matching the slower hole flux. Consequently, the EQE improvement is mechanistically explained by a dual effect: (1) the suppression of exciton quenching at the ETL/EML interface, and (2) the modification of charge accumulation at the interface through improved carrier balance.

To further improve device performance, researchers commonly employ the following strategies: incorporating an insulating interlayer to suppress electron leakage and block holes [28]; establishing a dipole layer to modulate the injection barrier [41]; or applying a passivation layer to mitigate surface defects in the ETL [34]. Here, a MgCl_2 interface intermediate layer (IML) is introduced, and primarily manifests in two aspects: firstly, leveraging its interfacial dipole effect to effectively suppress leakage current and block electrons; secondly, the Cl^- ions can precisely passivate surface defects, reducing non-radiative recombination losses. The QLEDs were fabricated by employing MgCl_2 as the IML (Fig. 3(a)). The corresponding cross-sectional TEM image of the QLED is depicted in Fig. 3(b). The optimized structure are: glass/ITO/PEDOT:PSS (25 nm)/TFB (20 nm)/QDs (~ 25 nm)/ MgCl_2 (~ 7 nm)/CSO (~ 23 nm)/Ag (100 nm). The energy level diagram of the device structure is shown in Fig. 3(c). Depth profiling XPS of Mg was conducted to detect the interface layer. The Mg 1s peaks appeared at about 23 nm depth after 97 s of

etching (Fig. S8 in the ESM). After 127 s etching, the characteristic peaks became barely detectable. According to the EDS analysis results (Fig. S9 in the ESM), both the ITO/QD and ITO/QD/ MgCl_2 samples exhibit uniformly distributed Cd and S signals, indicating the good film uniformity of the QD layers. After introducing MgCl_2 , additional Mg and Cl signals are clearly detected, and their relatively uniform distribution within the interfacial region suggests the formation of a uniform interfacial modification layer on the QD surface. The QLED devices with and without the MgCl_2 interlayer exhibit identical EL peaks at 630 nm (Fig. S10 in the ESM), indicating that the introduction of MgCl_2 does not alter the emission peak of the QDs or introduce any parasitic emission. Devices with MgCl_2 IML exhibit markedly reduced current density at low bias voltages (Fig. 3(d)), demonstrating effective suppression of both leakage currents and non-radiative recombination pathways. The efficacy of the MgCl_2 IML modification is further evidenced by the consistently higher CE values across all luminance compared to the control devices. The 5.0 wt.% CSO based QLED reached a significant enhancement in EQE, with a peak value increasing from 21.5% to 33.1% (Fig. 3(e)). Compared with CSO devices, the introduction of the MgCl_2 interlayer not only suppresses interfacial non-radiative recombination but also strengthens charge confinement at the QD/ETL interface. As a result, the MgCl_2 -modified devices maintain improved charge balance over a broader luminance range, partially extending the effective operating regime of the CSO strategy toward higher

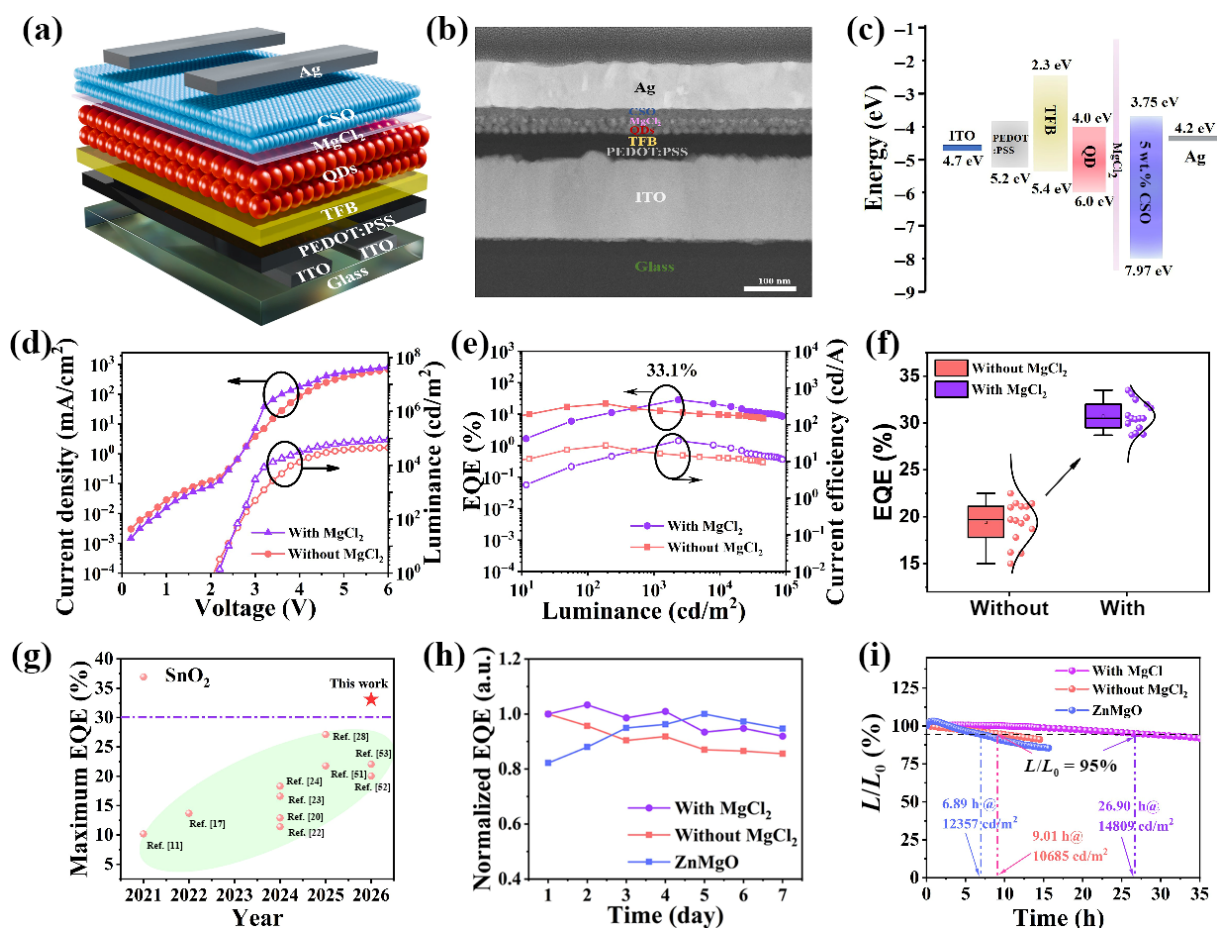


Figure 3 EL performances of QLEDs with or without MgCl_2 IML. (a) Structure diagram, (b) cross-section TEM image, (c) energy level diagram, (d) J - V - L curves, (e) EQE and CE as a function of luminance, and (f) EQE distributions measured from 25 devices. (g) Comparison of our maximum EQE with reported results. (h) The shelf stability measurements and (i) the device lifetimes.

brightness. To evaluate reproducibility, 25 QLEDs were fabricated and characterized by multiple batches, and an average EQE of 30.8% is obtained (Fig. 3(f)). To the best of our knowledge, so far, the QLEDs with MgCl_2 IML reached a record 33.1% EQE for SnO_2 -based QLEDs (Fig. 3(g) and Table 2), which is comparable to the state-of-the-art ZnMgO -based QLEDs [34, 42–49]. EL performance of QLEDs with different thickness IML is provided in Fig. S11 in the ESM. In addition, shelf stability is crucial critical parameter for evaluating the commercial viability of QLEDs. The CSO-based QLEDs effectively suppress the positive aging behavior (Fig. 3(h)), retaining 92% of their initial EQE after 7 days of storage. In contrast, the ZnMgO -based QLEDs exhibit a pronounced “increase-then-decrease” trend in EQE during the 7-day storage period, which is characteristic of the positive aging effect caused by interfacial instability and dynamic defect evolution. Unlike ZnMgO -based QLEDs, which exhibit a pronounced positive aging effect during lifetime testing, SnO_2 -based devices completely eliminate this phenomenon, indicating that the SnO_2 ETL is an ideal alternative to ZnMgO . The operational stability of QLEDs was evaluated under constant driving current (Fig. 3(i)). The QLEDs with MgCl_2 IML achieved a T_{95} lifetime of 26.90 h at an initial luminance (L) of $14,809 \text{ cd/m}^2$. With an acceleration factor of 1.792 (Fig. S12 in the ESM), T_{95} lifetimes were extrapolated to 3367.7 h at 1000 cd/m^2 , which is 5.36-fold higher than the control one. These results further demonstrate the significant advantage of the SnO_2 -based QLEDs for achieving stable QLEDs.

Figure 4(a) shows the high-resolution XPS spectra of the O 1s peaks for 5.0 wt.% CSO film with or without MgCl_2 IML. The observed decrease in O_H (9.37% to 6.85%) and O_V (31.84% to 22.08%) content is attributed to the effective passivation by MgCl_2 , signaling a marked decrease in both exciton recombination centers and interfacial defects. At the same time, Raman testing was conducted (Fig. S13 in the ESM), and the results showed the Raman spectra of 5 wt.% Co- SnO_2 and MgCl_2 -modified samples exhibit two characteristic peaks at approximately 576 and 625 cm^{-1} ,

which are attributed to the O–Sn–O bending vibration and Sn–O stretching vibration modes, respectively [29]. After MgCl_2 treatment, both peaks show a slight blue shift accompanied by a noticeable reduction in the FWHM of the 576 cm^{-1} peak. These changes indicate enhanced lattice ordering and reduced phonon scattering, suggesting a more uniform Sn–O bonding environment. The results imply a decreased concentration of oxygen vacancy-related defects. This behavior can be attributed to the passivation of oxygen vacancies by Cl^- ions, likely through surface coordination or charge redistribution, rather than direct lattice substitution.

As PL decay patterns shown in Fig. 4(b), the τ_{avg} of sample with MgCl_2 IML is 17.02 ns , a substantial increase from the 15.0 ns of QDs/CSO. To further clarify the role of MgCl_2 , additional QD/ MgCl_2 control samples were prepared and characterized. Compared with pristine QDs, the QD/ MgCl_2 samples exhibited enhanced PLQY and prolonged PL lifetime (Fig. S14 and Table S6 in the ESM), confirming that the MgCl_2 interfacial layer effectively passivates surface defects and suppresses exciton quenching. Based on the measured PLQY and τ_{avg} , the radiative recombination rate (k_r) and non-radiative recombination rate (k_{nr}) were further calculated (Table S7 in the ESM). After MgCl_2 treatment, k_{nr} significantly decreased from $0.03277 \times 10^{-9}/\text{s}$ to $0.02238 \times 10^{-9}/\text{s}$, indicating that the improved PL intensity and prolonged lifetime mainly originate from the suppression of defect-assisted non-radiative recombination. Consequently, sample with MgCl_2 IML exhibits a stronger steady-state PL intensity (Fig. S15 in the ESM). The XPS depth profiles of Mg^{2+} and Cl^- recorded from a CSO/ MgCl_2 /QDs/TFB film are shown in Fig. 4(c), and a fitted 284 eV peaks come from C–C/C–H of the QD ligands (Fig. S16 in the ESM). The disappearance of Cl^- peaks is delayed by 15 s after Mg^{2+} , indicating the passivation effect of Cl^- on QDs. To elucidate the role of the Cl^- , the energy band structure of a QD/ MgCl_2 film was characterized. From the Tauc plot in Fig. S17 in the ESM, the QDs/ MgCl_2 film exhibits an E_g of 1.98 eV . As UPS shown in Fig. 4(d), the measured VBM and CBM moved up ~ 0.27 and

Table 2 Summary of the used QD, device structure, peak EL wavelength, maximum EQE (EQE_{max}), lifetime of SnO_2 -based QLEDs

Year	QD	Structure	EL (nm)	$\text{EQE}_{\text{max}}/\text{CE}_{\text{max}}$	Lifetime (h)	Ref.
2020	CdSe	ITO/ SnO_2 /QD/CBP/ MoO_3 /Al	Red	11.8 cd/A	$T_{50}(@1700 \text{ cd/m}^2) = 5.1$	[19]
2021	CdSe/ZnS	ITO/ SnO_2 /QD/CBP/TCTA/TAPC/ MoO_3 /Ag	632	10.18%	$T_{50}(@500 \text{ cd/m}^2) = 2168$	[11]
2022	CdSe/ZnS	ITO/ SnO_2 /QD/CBP/ MoO_3 /Al	Red	6.78 cd/A	—	[15]
2022	CdZnSeS/ZnS	ITO/ SnO_2 @QD/CBP/ MoO_3 /Al	518	9.42%	$T_{50}(@100 \text{ cd/m}^2) = 700$	[54]
2022	CdSe/ $\text{Cd}_{1-x}\text{Zn}_x\text{Se}$ /ZnSe	ITO/PEDOT:PSS/TFB/QD/TMAH- SnO_2 /Ag	625	13.7%	$T_{95}(@1000 \text{ cd/m}^2) = 3200$	[17]
2023	CdSe/ZnS	ITO/SVA- SnO_2 /QD/CBP/ MoO_3 /Al	Red	10.8 cd/A	—	[18]
2024	CdSe/ZnSe/ZnS	ITO/ SnO_2 /QD/CBP/ MoO_3 /Al	Red	12.9%	$T_{50}(@1436 \text{ cd/m}^2) = 1.68$	[20]
2024	CdSe	ITO/PEDOT:PSS/TFB/QD/Mn- SnO_2 /Al	631	11.4%	$T_{95}(@1000 \text{ cd/m}^2) = 1009.2$	[22]
2024	CdSe/ZnSe/ZnS	ITO/PEDOT:PSS/TFB/QD/Zr- SnO_2 /Al	638	16.6%	—	[23]
2024	ZnCdSeS	ITO/PEDOT:PSS/TFB/QD/Sb- SnO_2 /Ag	630	18.3%	$T_{95}(@1000 \text{ cd/m}^2) = 4320$	[24]
2024	CdSe/ZnS	ITO/PEDOT:PSS/PVK:CBP/QD/Zn- SnO_2 /Al	Green	5.03%	$T_{50}(@100 \text{ cd/m}^2) = 9141$	[21]
2025	CdSe/ZnSe/ZnS	ITO/PEDOT:PSS/TFB/QD/ CaMoO_4 /Zn- SnO_2 /Al	627	27.1%	$T_{95}(@1000 \text{ cd/m}^2) = 1828.3$	[28]
2025	CdSe/ZnSe/ZnS	ITO/In- SnO_2 /QD/CBP/ MoO_3 /Al	Red	21.79%	—	[51]
2026	CdSe/ZnSe/ZnS	ITO/Ga- SnO_2 /QDs/CBP/ MoO_3 /Al	Red	20.04%	—	[52]
2026	CdSe/ZnSe/ZnS	ITO/Mg- SnO_2 /QDs/CBP/ MoO_3 /Al	627	22.04%	$T_{50}(@1049 \text{ cd/m}^2) = 4.2$	[53]
2026	CdSe/ZnS	ITO/PEDOT:PSS/TFB/QD/ MgCl_2 /Co- SnO_2 /Ag	630	33.1%	$T_{95}(@1000 \text{ cd/m}^2) = 3367.7$	This work

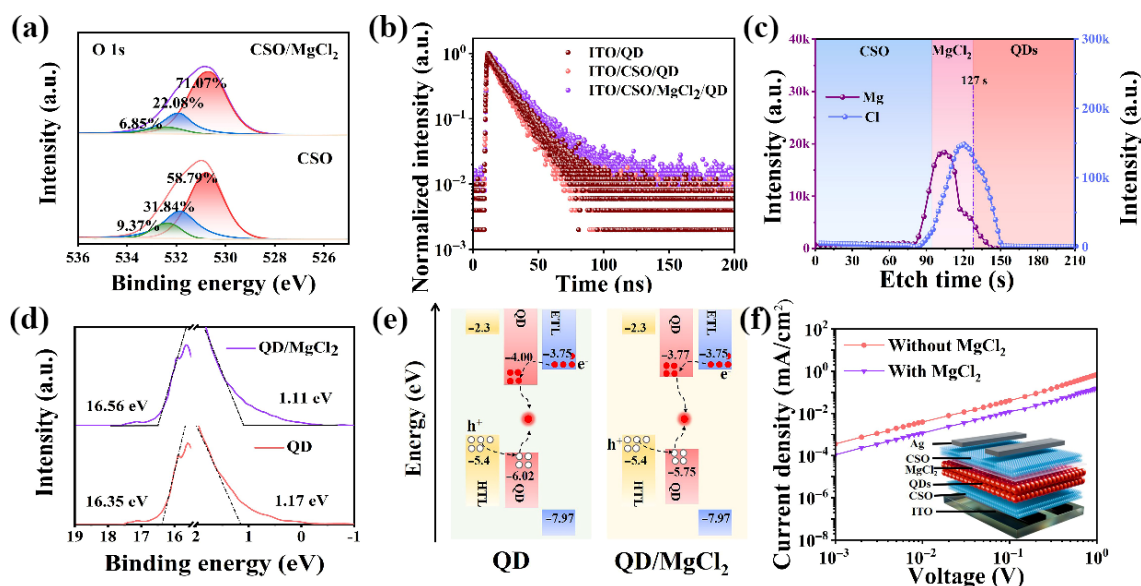


Figure 4 (a) High-resolution XPS spectra of O 1s orbit of 5 wt.% CSO film, (b) TRPL spectra of ITO/CSO/QDs sample, (c) XPS depth profiling, (d) UPS spectra, (e) energy level diagram, and (f) J - V curve of EODs with or without MgCl₂ IML.

~ 0.23 eV, respectively. The UPS results suggest that MgCl₂ induces local interfacial electronic structure modulation near the QD/CSO interface. Considering the ultrathin nature of the MgCl₂ interlayer, the observed energy-level variation is more likely associated with local vacuum-level shift and interfacial band bending rather than an overall intrinsic energy-level shift of the entire QD layer (Fig. 4(e)). Given that hole mobility in TFB [50] is an order of magnitude lower than electron mobility (Table S3 in the ESM), the MgCl₂ modification balances carrier injection, mitigates hole accumulation and suppresses nonradiative recombination, leading to more efficient radiative recombination across the QD layer. Besides, the RMS roughness of the ITO/QDs/MgCl₂ film decreased from 2.129 to 2.109 nm (Fig. S18 in the ESM), showing a smoother film and attribute to lower leakage current. As shown in Fig. S19 in the ESM, the Nyquist plots of QLEDs exhibit a semicircular shape, which stems from those inherent resistor-capacitor (RC) circuit configuration. This result confirms that the MgCl₂ IML enhances the overall complex resistance of the LED devices. The EODs with or without the MgCl₂ were shown in Fig. 4(f). The electrical characterization reveals that the MgCl₂ IML effectively moderates electron transport, thereby achieving better charge balance. In addition, hole-only devices incorporating the MgCl₂ IML exhibited enhanced hole current density (Fig. S20 in the ESM), suggesting that MgCl₂ can also facilitate hole injection. To gain deeper insight into the role of the MgCl₂ IML in regulating carrier dynamics, time-resolved EL (TREL) measurements were performed. As seen in Fig. S21 in the ESM, the QLEDs with MgCl₂ IML show a longer rise time than devices without MgCl₂ IML counterpart. This delayed rise time is more likely associated with the regulated electron injection dynamics and interfacial charge redistribution induced by the MgCl₂ interlayer. The ultrathin MgCl₂ layer together with the local interfacial dipole effect can retard electron accumulation and injection at the QD/ETL interface, resulting in a slower EL establishment process.

Overall, the improved device performance is primarily attributed to the defect-passivation effect of MgCl₂. The reduced oxygen-defect-related signal intensity in XPS together with the suppressed nonradiative recombination revealed by PLQY/TRPL

measurements indicates that Cl⁻ effectively passivates interfacial trap states and suppresses exciton quenching at the QD/CSO interface. In addition, MgCl₂ may also induce local interfacial dipoles and electric-field redistribution, which further regulate electron injection dynamics and optimize charge balance. The synergistic effects collectively contribute to the enhanced efficiency and stability of the QLED devices.

4 Conclusion

In summary, we synthesized well-dispersed CSO as an electron transport layer with reduced oxygen vacancies and surface hydroxyl groups. The improved interfacial properties effectively suppress exciton quenching and enable a peak EQE of 21.5% for red QLEDs at a luminance of 187 cd/m². However, at high luminance the carrier-balance improvement cannot be maintained and the CSO-device still shows significant efficiency roll-off. To further suppress exciton quenching at the QD/ETL interface, an ultrathin MgCl₂ interlayer was introduced, which simultaneously passivates interfacial defects, optimizes energy-level alignment, and improves film quality. The MgCl₂-based QLED achieves a maximum EQE of 33.1% and realizes enhanced performance over a wider operating range. A 5.36-fold increased operational lifetime is obtained for the best performance MgCl₂-based QLED with a T_{95} of 3367.7 h. This work demonstrates that the combination of bulk defect regulation and interface engineering effectively mitigates exciton quenching and carrier imbalance in SnO₂-based QLEDs, offering a practical strategy for achieving highly efficient and operationally stable devices.

Electronic Supplementary Material: Supplementary material (the full-scan XPS Spectra of CSO; XPS peak-fitting results of oxygen species in SnO₂; PL spectra of different CSO NPs; AFM images of SnO₂ NPs and 5.0 wt.% CSO; J - V characteristics of EODs with different ETLs; electron mobility of SnO₂ with different Co doping concentrations; summary of the EL Performance of QLEDs with Different ETLs; PL decay parameters of QDs in different samples; 2D Mg²⁺ XPS depth profiles; normalized EL spectra of QLEDs with

or without MgCl_2 IML; EL performance of QLEDs with different concentrations of MgCl_2 interlayer.; T_{95} lifetime as a function of initial luminance for the QLED with MgCl_2 interlayer; the shelf stability measurements; PL spectra of ITO/CSO/QDs; Tauc plot of QD film and QD/ MgCl_2 film; AFM image of QD film and QD/ MgCl_2 film; Nyquist curves and TREL spectra) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909019>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

L. S. W.: Conceptualization, data curation, formal analysis, funding acquisition, methodology, visualization, writing – original draft, writing – review & editing. G. F. L.: Data curation, formal analysis, investigation, methodology, visualization, writing – review & editing. R. J. X. and D. C. P.: Data curation, formal analysis. H. H. W., Z. R. H., S. Y. H., Z. B. T. and X. Y. G.: Data curation. H. Z.: Data curation, visualization. J. L. Z. and B. S. Z.: Funding acquisition, visualization, resources, writing – review & editing. All the authors have approved the final manuscript.

Use of AI statement

None.

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