Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.


# Supporting Information

# Mixed-Dimensional 0D/3D Perovskite Heterostructure for Efficient Green Light Emitting Diodes

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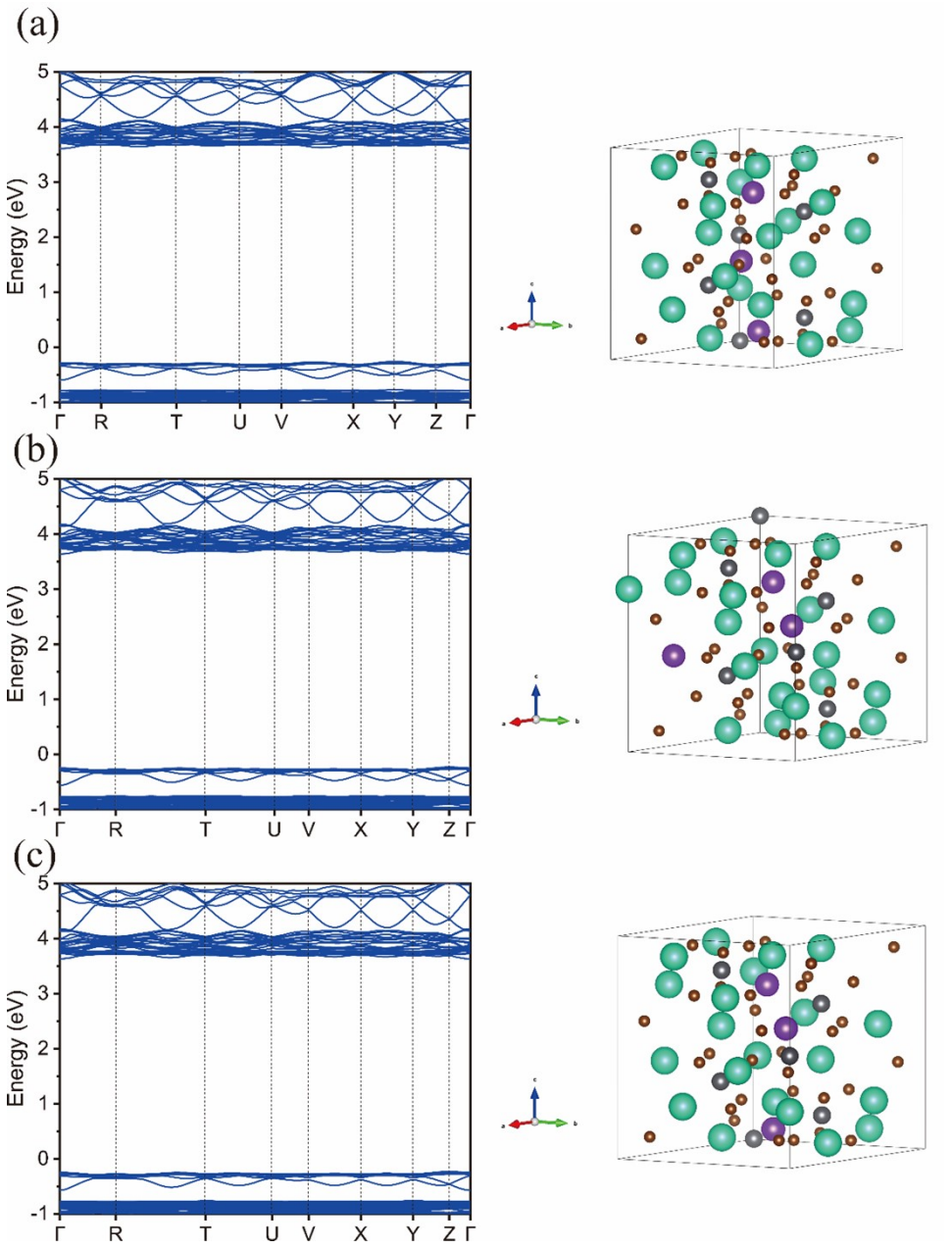


**Figure S1** Energy band structure of the sample with three Cs atoms are replaced by K atoms, denoted as $Cs_{21}K_3Pb_6Br_{36}$. The three different K positions (a), (b), (c) are considered for the $Cs_{21}K_3Pb_6Br_{36}$.

It indicates that the K incorporation has little effect on the bandgap shift. The 0D $Cs_{4-x}K_xPbBr_6$ also has a large bandgap that can form the 0D/3D heterostructure for facilitating the radiative recombination.

The formation energies of K incorporated were calculated by the following formulas:

$E_{form}$ = E(incorporated) - E(pure) - $\mu$(K) + $\mu$(Cs)

where E(incorporated) and E(pure) are the total energies of K- incorporated and pure $Cs_4PbBr_6$, respectively. $\mu$(K) and $\mu$(Cs) are chemical potentials of K or Cs atom, respectively.

The chemical potentials of K or Cs atom

K: -2.0589750/2 = -1.0294875 eV

Cs: -1.7039335/2 = -0.85196675 eV

$Cs_4PbBr_6$: -207.47095 eV

1-K-$Cs_4PbBr_6$: -207.32740 eV

3-K-$Cs_4PbBr_6$ -207.06845 eV

The Formation energies of K incorporated：

1-K-$Cs_4PbBr_6$:

-207.32740 + 207.47095 + 1.0294875 - 0.85196675 = 0.32107075 eV

3-K-$Cs_4PbBr_6$

-207.06845+ 207.47095 + 3×1.0294875 - 3×0.85196675 = 0.93506225 eV

The average formation energy of K：0.312 eV, the average foramtion energy confirm the possibality for the formation of $Cs_{4-x}K_xPbBr_6$.

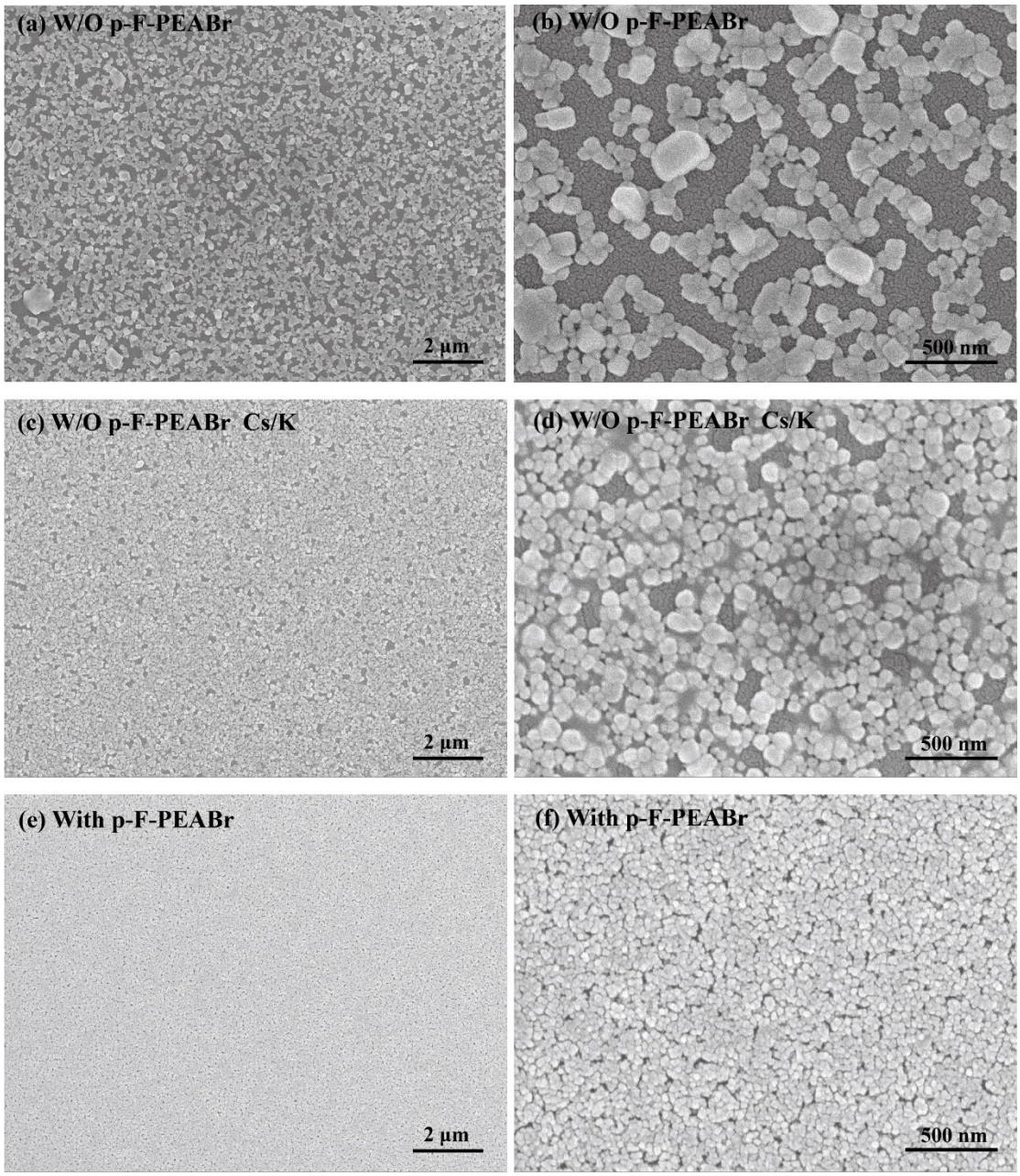


**Figure S2** FE-SEM of (a) low and (b) high magnification of the $CsPbBr_3$ films, (c) low (d) high magnification of the $(Cs/K)PbBr_3$ films, (e) low and (f) high magnification of the p-F-PEABr treatment $CsPbBr_3$ films.

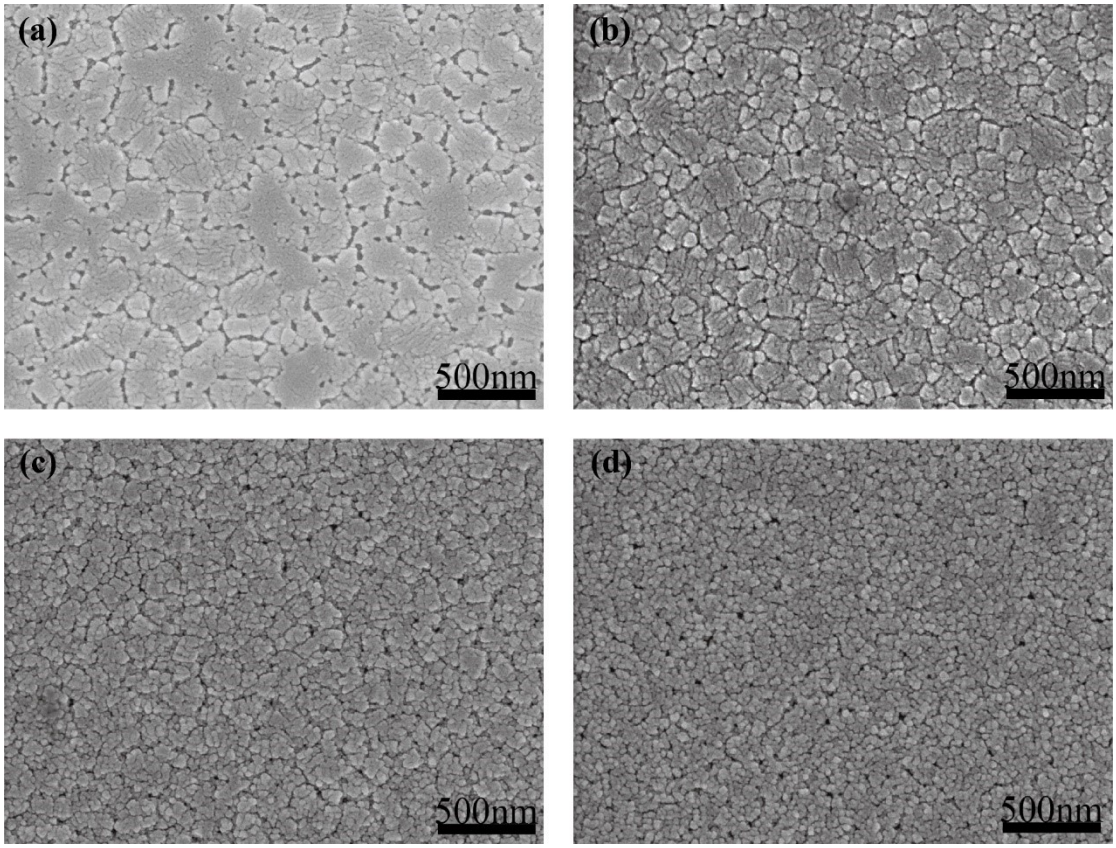


**Figure S3** FE-SEM of (a) p-F-PEABr, (b) p-F-PEABr DMSO:DMF=4:1, (c) p-F-PEABr DMSO:DMF=4:1 PEO, (d) p-F-PEABr DMSO:DMF=4:1 PEO 4 mg/ml CE 2 mg/ml, scale bar 500 nm. The samples were prepared with the molar ratios of 1.1:1:0.22, for CsBr:$PbBr_2$:p-F-PEABr.

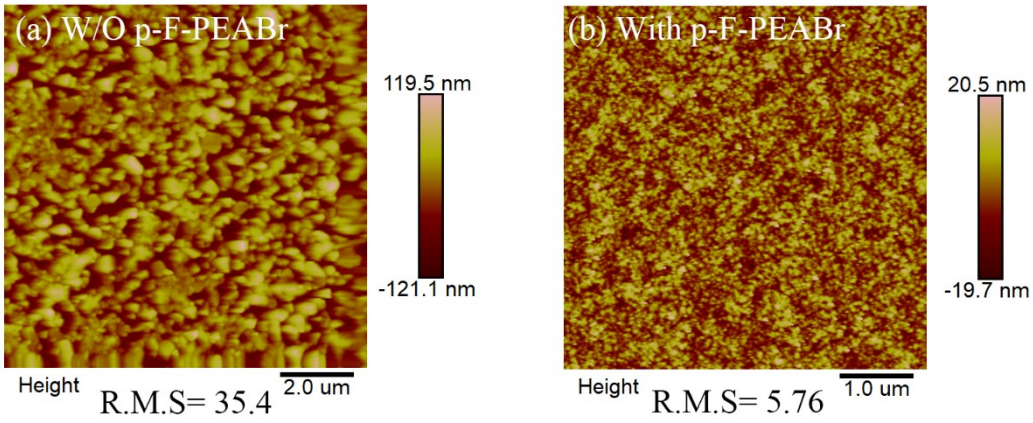


**Figure S4** The atomic force microscopy (AFM) images show that the morphology of the perovskite film (a) without p-F-PEABr treatment and (b) with p-F-PEABr treatment. The samples were prepared with the molar ratios of 1:1:0 and 1:1:0.2, respectively, for CsBr:$PbBr_2$:p-F-PEABr.

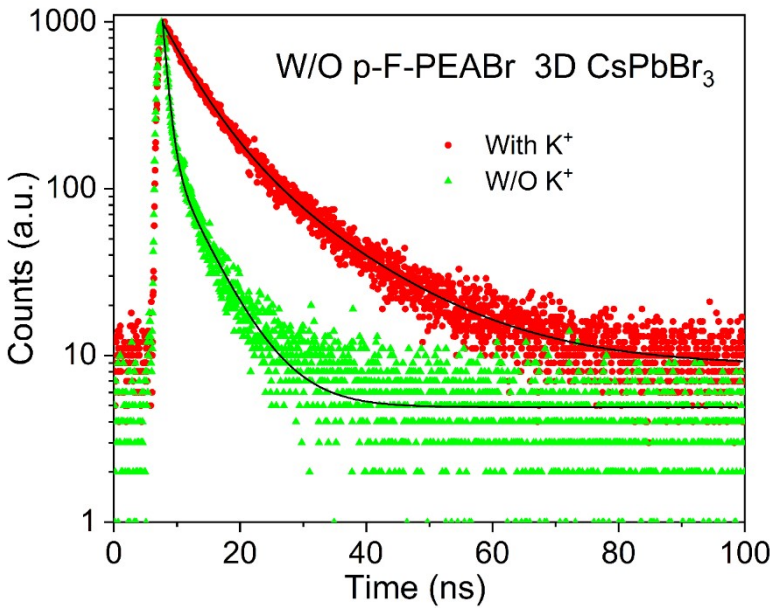


**Figure S5** TRPL of the 3D $CsPbBr_3$ with and without $K^+$ addition. The samples were prepared with the molar ratios of 1.1:1:0 and 1.1:1:0.05, for $CsBr:PbBr_2:KBr$ and dissolved in pure DMSO.

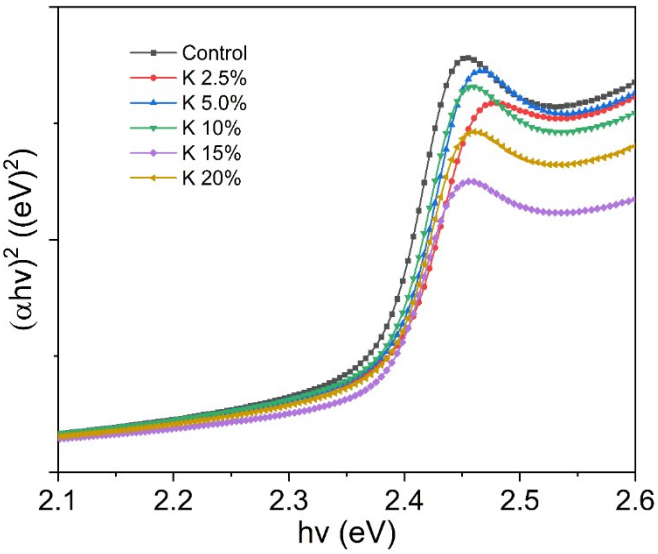


**Figure S6** Tauc plots showing the dependence of $(\alpha h\nu)^2$ of 3D $CsPbBr_3$ and 0D/3D heterostructure perovskite films upon the incident photon energy $(h\nu)$ (assuming direct allowed transitions).

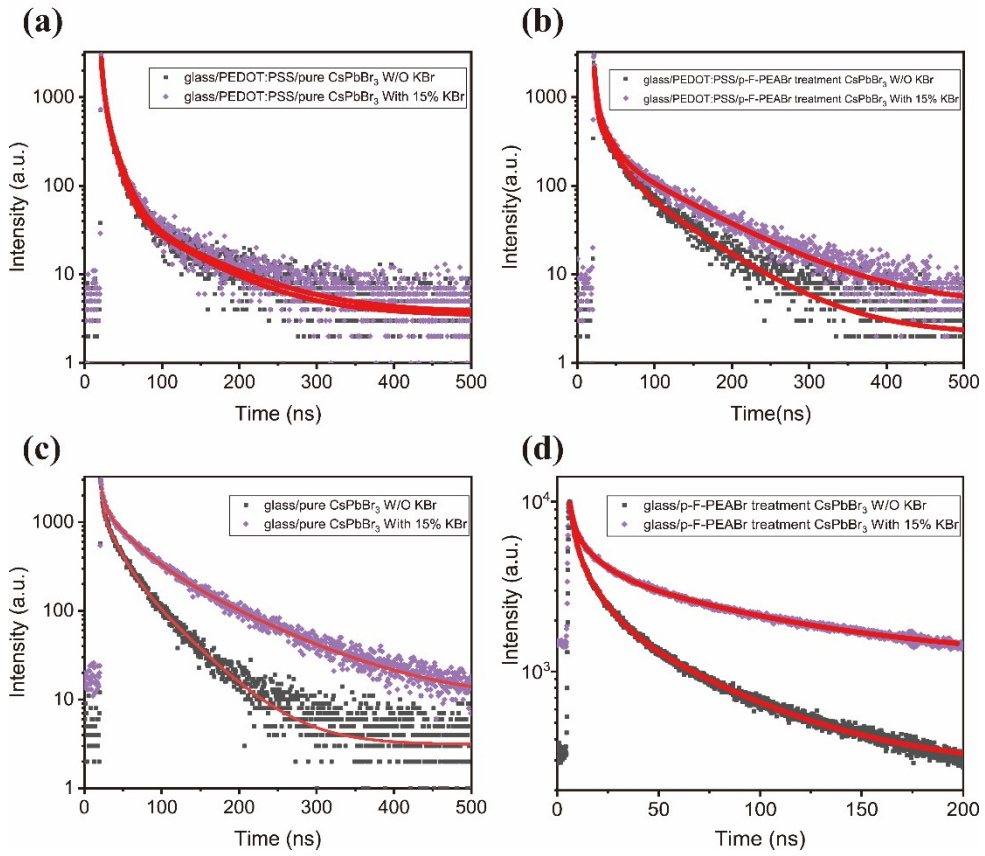


**Figure S7** TRPL decay curves of (a) 3D $CsPbBr_3$ and K15% 0D/3D heterostructure perovskite and (b) p-F-PEABr treatment 3D $CsPbBr_3$ and K15% 0D/3D heterostructure perovskite with the configuration of glass/PEDOT:PSS/perovskite films. (The perovskite were dissloved in DMSO/DMF mixture solvent (4:1, by volume) along with PEO 4mg/ml CE 2mg/ml) (c) 3D $CsPbBr_3$ and K15% 0D/3D heterostructure perovskite and (d) p-F-PEABr treatment 3D $CsPbBr_3$ and K15% 0D/3D heterostructure perovskite with the configuration of glass/perovskite films.

The fitting results are listed in Tables S3 and S4.

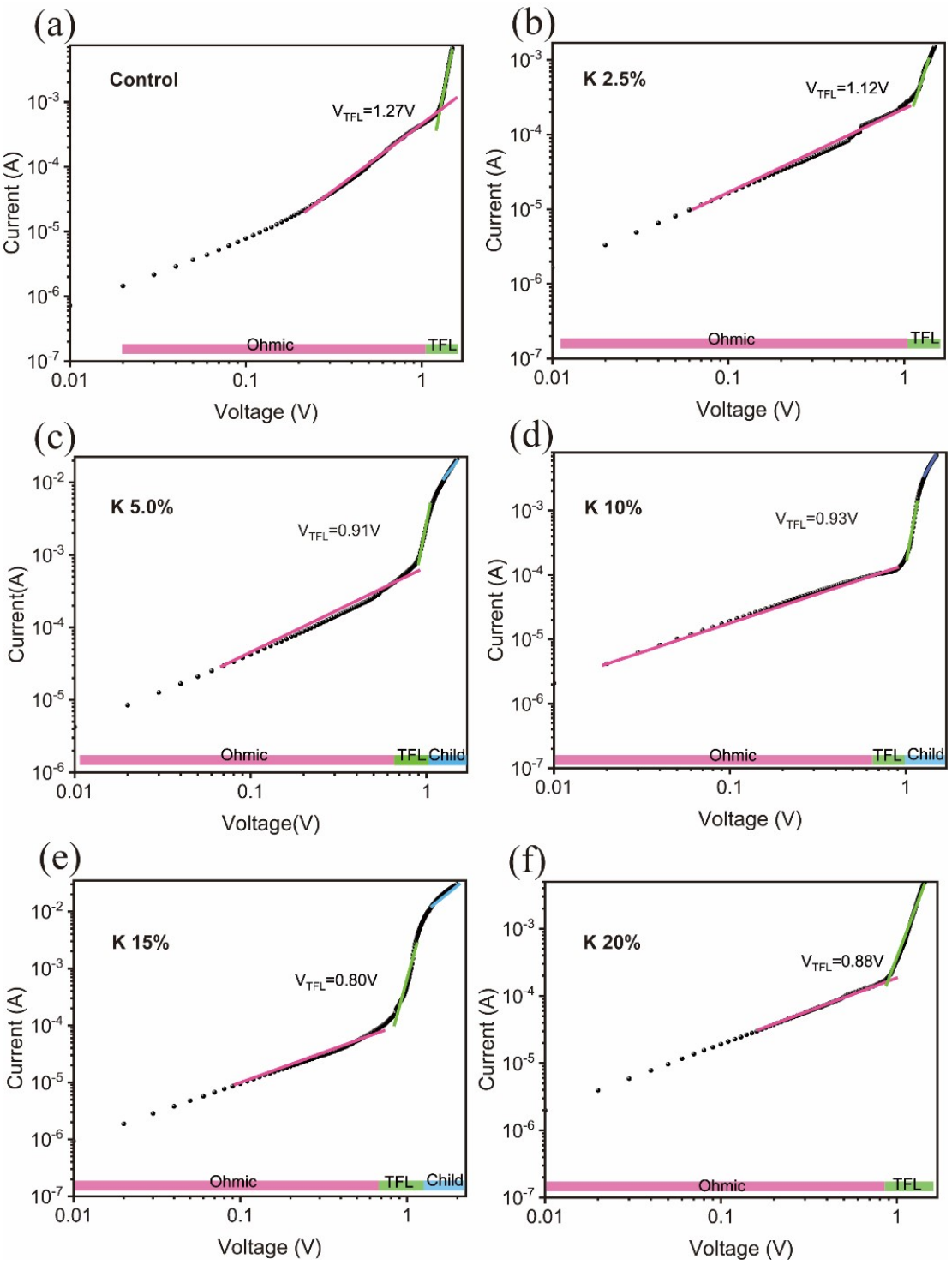


**Figure S8** Dark current-voltage measurements of the hole only device, with the structure of ITO/PEDOT:PSS/perovskite/$MoO_3$/Ag with various $K^+$ addition for 3D $CsPbBr_3$ and 0D/3D heterostructure perovskite.

The trap density $(n_t)$ of the perovskite thin films with different of $K^+$ addition can be extracted by the equation $n_t = \frac{2V_{TFL}\varepsilon\varepsilon_0}{eL^2}$, where $\varepsilon_0$, $e$ and $L$ represent vacuum permittivity, elementary charge and thickness of the perovskite film, respectively; $\varepsilon$ is the average relative dielectric constant of the perovskite thin films with different potassium ratios. The red line represents the ohmic regime of each case, and the green line indicates the trap-filled limit (TFL) regime with the onset voltage ($V_{TFL}$). Considering the different perovskite films were spin-coated with the same process, we assume the same film thickness. Generally, the lower value of $V_{TEF}$ indicates the lower concentration of trap states in the film.

$\varepsilon_0$ is the vacuum permiittivity ($8.854\times10^{-12}$ F $m^{-1}$)

$\varepsilon$ is the relative dielectric constant of the perovskite layer; we tend to take it as 19.2 from the previous report.[1]

$L$ is the thickness of the perovskite emitter layer (taken as 50 nm from the SEM result)

| Samples | $\eta_t/cm^{-3}$ |
|---|---|
| Control | $1.079\times10^{18}$ |
| K 2.5 | $9.520\times10^{17}$ |
| K 5.0 | $7.735\times10^{17}$ |
| K 10 | $7.905\times10^{17}$ |
| K 15 | $6.800\times10^{17}$ |
| K 20 | $7.480\times10^{17}$ |

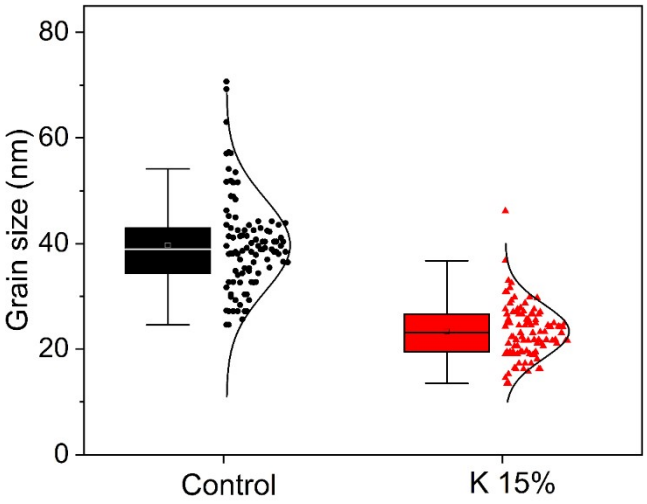


**Figure S9** Particle size distribution of the p-F-PEABr treatment 3D $CsPbBr_3$ for control and K15% 0D/3D heterostructure perovskite samples.

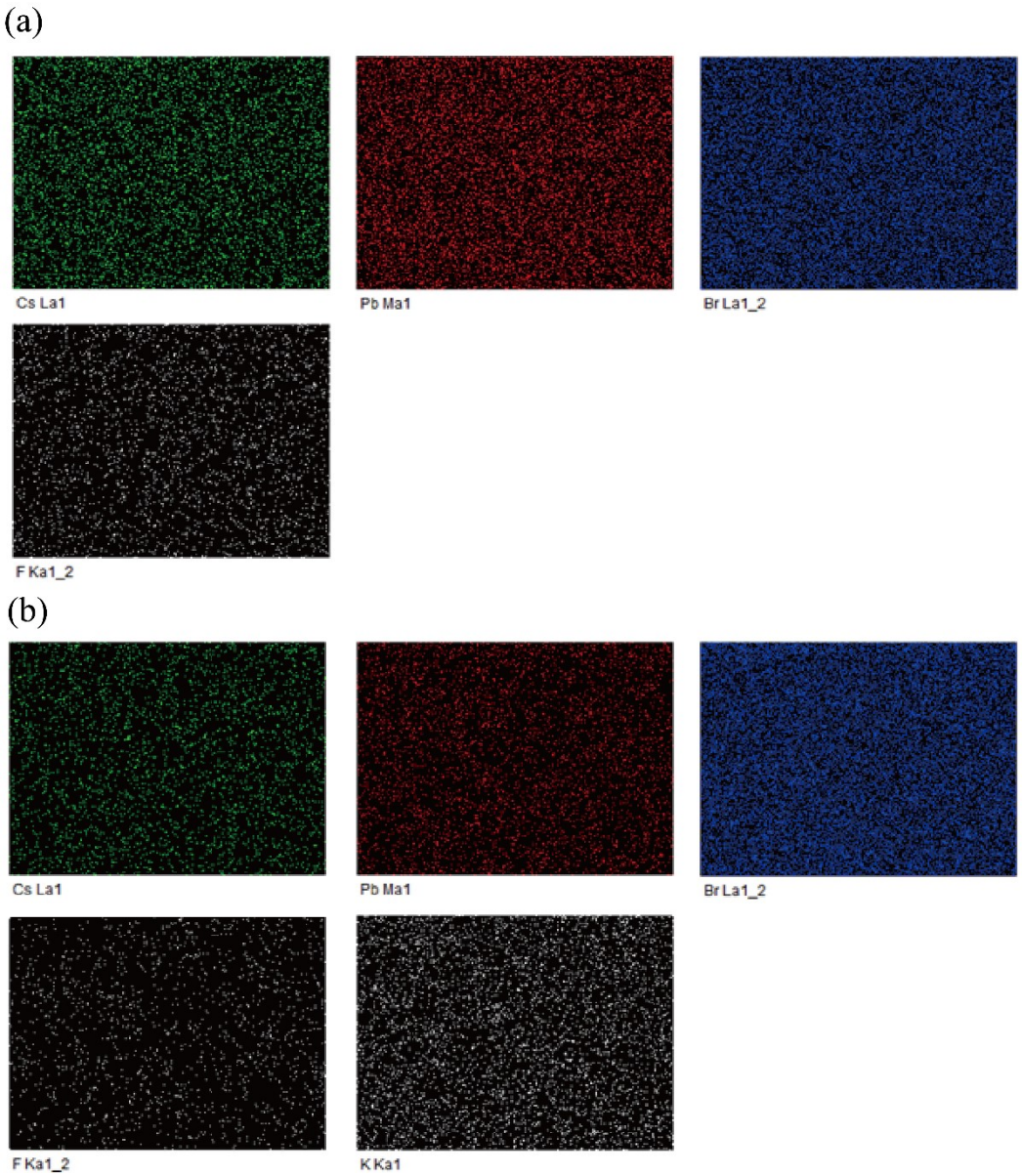


**Figure S10** Elemental mapping of the perovskite films showing the distribution of the homogeneous elements of (a) the p-F-PEABr treatment 3D $CsPbBr_3$ and (b) the p-F-PEABr treatment K15% 0D/3D heterostructure perovskite

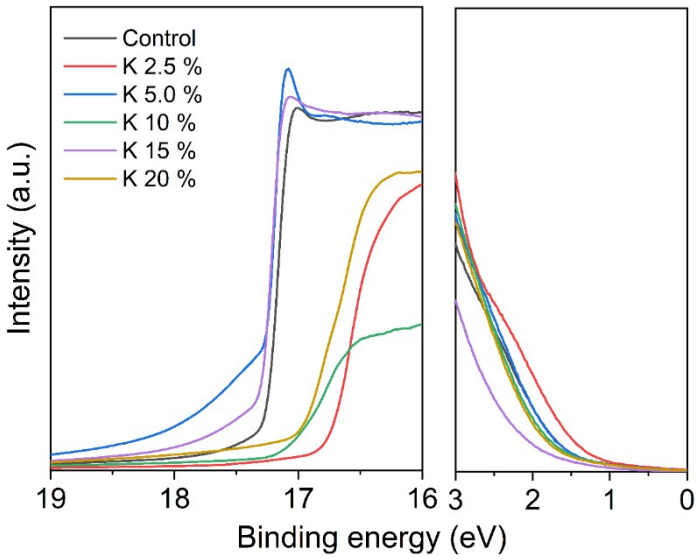


**Figure S11** UPS Spectra of the p-F-PEABr treatment 3D $CsPbBr_3$ and 0D/3D heterostructure perovskite showing the binding energy secondary-electron cutoffs (left) and HOMO regions (right).

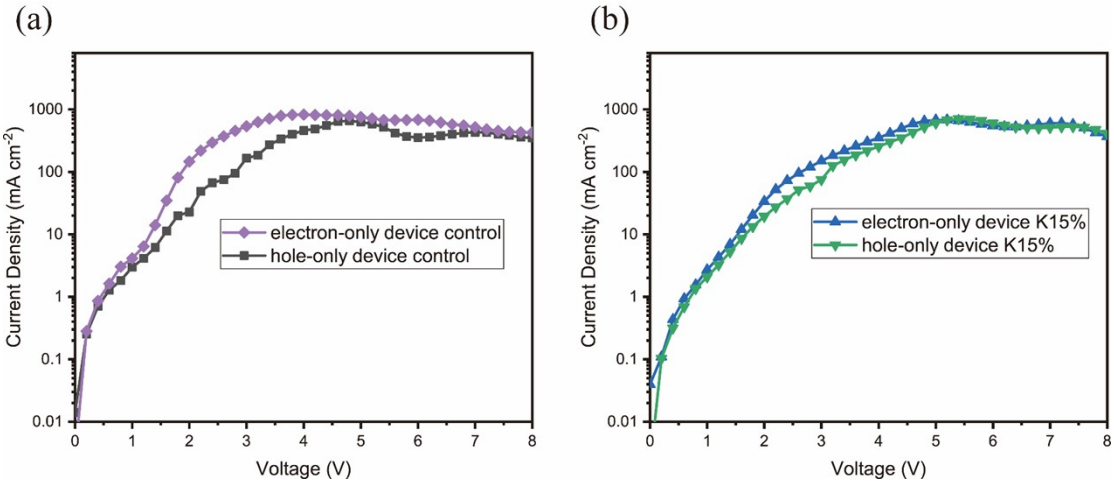


**Figure S12** Current density-voltage (J-V) curves of electron-only (ITO/ $SnO_2$/ perovskite/ TPBi/ LiF/ Al) and hole-only (ITO/ PEDOT:PSS/ perovskite/ $MoO_3$/ Ag) devices for (a) 3D $CsPbBr_3$ and (b) K15% 0D/3D heterostructure perovksite films.

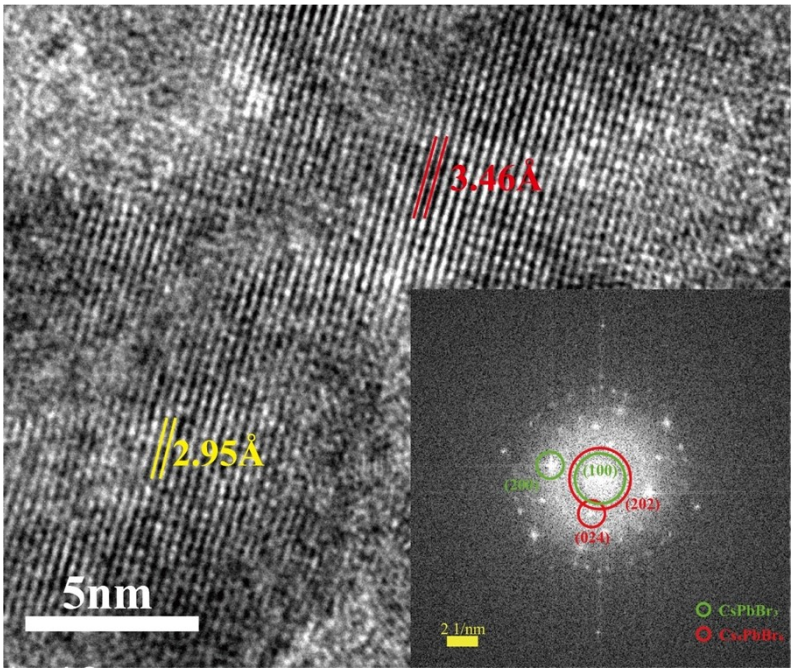


**Figure S13** HRTEM of the K20% 0D/3D heterostructure perovskite films with the corresponding FFT result.

**Table S1** Summary of the time-resolved photoluminescence lifetime fitting parameters of the 3D $CsPbBr_3$ and 0D/3D heterostructure perovskite films.

| Samples | $\tau_1/ns$ | $A_1$ | $\tau_2/ns$ | $A_2$ | $\tau_3/ns$ | $A_3$ | $\tau_{avg}/ns$ |
|---|---|---|---|---|---|---|---|
| Control | 2.54 | 6.50 | 14.07 | 26.68 | 66.87 | 66.82 | 63.51 |
| K 2.5 | 2.37 | 6.41 | 13.00 | 24.62 | 71.50 | 68.98 | 66.10 |
| K 5.0 | 2.94 | 5.50 | 15.39 | 25.22 | 71.24 | 69.27 | 65.87 |
| K 10 | 2.37 | 6.63 | 12.73 | 22.54 | 75.00 | 70.83 | 68.18 |
| K 15 | 2.57 | 2.58 | 14.85 | 12.41 | 99.36 | 85.01 | 83.40 |
| K 20 | 3.42 | 3.40 | 17.80 | 16.55 | 88.12 | 80.05 | 77.39 |

**Table S2** Summary of the fitting results of the PL temperature dependence for the control and K15% 0D/3D heterostructure perovskite samples. (Based on the combined exciton-trap model)

| Sample | $D_1$ | $E_q/meV$ | $C_1$ | $E_j/meV$ | I(0) |
|---|---|---|---|---|---|
| Control | 47.22 | 38.33 | 117.41 | 56.36 | $2.94 \times 10^7$ |
| K15% 0D/3D heterostructure | 7.11 | 8.34 | 37.04 | 64.20 | $1.22 \times 10^7$ |

**Table S3** Summary of the time-resolved photoluminescence lifetime fitting parameters of the glass/PEDOT:PSS/ Pure $CsPbBr_3$ perovskite films with and without 15% KBr, glass/PEDOT:PSS/p-F-PEABr treatment 3D $CsPbBr_3$ and K15% 0D/3D heterostructure perovskite films.

| Samples | $\tau_1/ns$ | $A_1$ | $\tau_2/ns$ | $A_2$ | $\tau_3/ns$ | $A_3$ | $\tau_{avg}/ns$ |
|---|---|---|---|---|---|---|---|
| Pure $CsPbBr_3$ W/O $K^+$ | 2.58 | 24.17 | 11.23 | 52.86 | 75.56 | 22.97 | 23.92 |
| Pure $CsPbBr_3$ With 15% K | 3.32 | 25.00 | 14.08 | 52.36 | 84.97 | 22.64 | 27.44 |
| p-F-PEABr treatment 3D $CsPbBr_3$ | 1.91 | 8.47 | 16.81 | 37.91 | 50.26 | 53.62 | 33.48 |
| p-F-PEABr treatment K15% 0D/3D heterostructure | 4.81 | 6.23 | 32.42 | 32.92 | 99.21 | 60.85 | 71.34 |

**Table S4** Summary of the time-resolved photoluminescence lifetime fitting parameters of the glass/3D $CsPbBr_3$ perovskite films with and without 15% KBr, glass/ p-F-PEABr treatment 3D $CsPbBr_3$ and K15% 0D/3D heterostructure perovskite films.

| Samples | $\tau_1/ns$ | $A_1$ | $\tau_2/ns$ | $A_2$ | $\tau_3/ns$ | $A_3$ | $\tau_{avg}/ns$ |
|---|---|---|---|---|---|---|---|
| 3D $CsPbBr_3$ W/O $K^+$ | 2.18 | 14.55 | 19.01 | 35.38 | 72.88 | 50.07 | 43.54 |
| 3D $CsPbBr_3$ With 15% K | 2.57 | 11.44 | 15.00 | 27.29 | 91.57 | 61.27 | 60.49 |
| p-F-PEABr treatment 3D $CsPbBr_3$ | 2.54 | 6.50 | 14.07 | 26.68 | 66.87 | 66.82 | 63.51 |
| p-F-PEABr treatment K15% 0D/3D heterostructure | 2.57 | 2.58 | 14.85 | 12.41 | 99.36 | 85.01 | 83.40 |

(1) Schlaus, A. P.; Spencer, M. S.; Miyata, K.; Liu, F.; Wang, X.; Datta, I.; Lipson, M.; Pan, A.; Zhu, X. Y. How lasing happens in $CsPbBr_3$ perovskite nanowires. *Nature Commun.* **2019,** *10* , 265.