Dimensionality Controls Anion Intermixing in Electroluminescent Perovskite Heterojunctions

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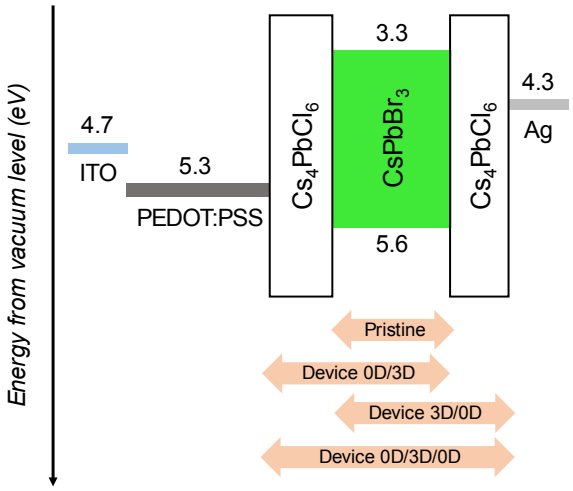

Figure S1. Schematic flat band energy diagram of the materials used in light-emitting devices based on Cs_4PbCl_6/$CsPbBr_3$ heterojunctions. Values for $CsPbBr_3$ are from the literature.[1] The reported IE for the iodide compound Cs_4PbI_6 is 7.15 eV;[2] considering a downward shift of the valence band maximum (VBM) when exchanging I for Cl (due to increased electronegativity),[3] we expect the IE of Cs_4PbCl_6 to be approximately 7.5 eV. Taking into account an optical bandgap of 4.4 eV, the electron affinity (EA) of Cs_4PbCl_6 would be of about 3 eV.

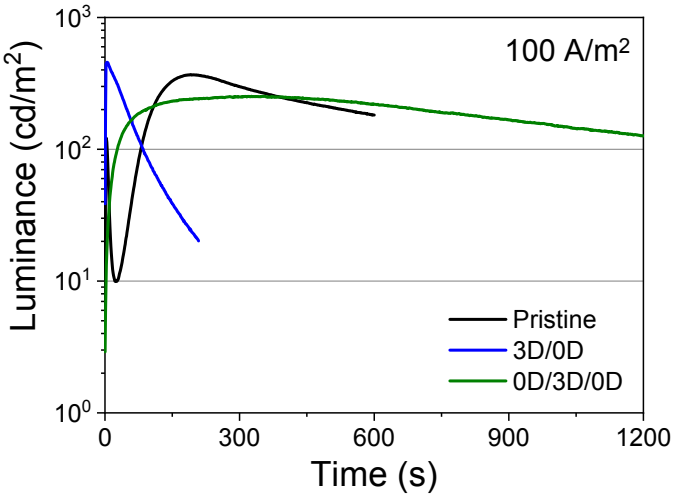

Figure S2. Constant current driving with $CsPbBr_3$ (Pristine), $CsPbBr_3/Cs_4PbCl_6$ (Device 3D/0D) $Cs_4PbCl_6/CsPbBr_3/Cs_4PbCl_6$ (Device 0D/3D/0D).

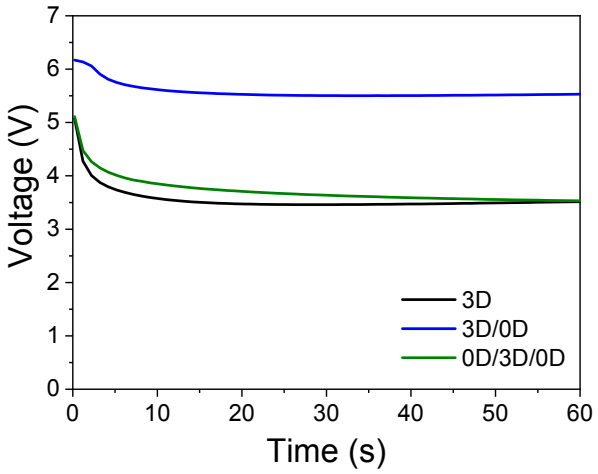

Figure S3. Zoom of the initial voltage decay upon application of a constant current of 100 A/m^2 to the light-emitting devices based on $Cs_4PbCl_6/CsPbBr_3$ heterojunction.

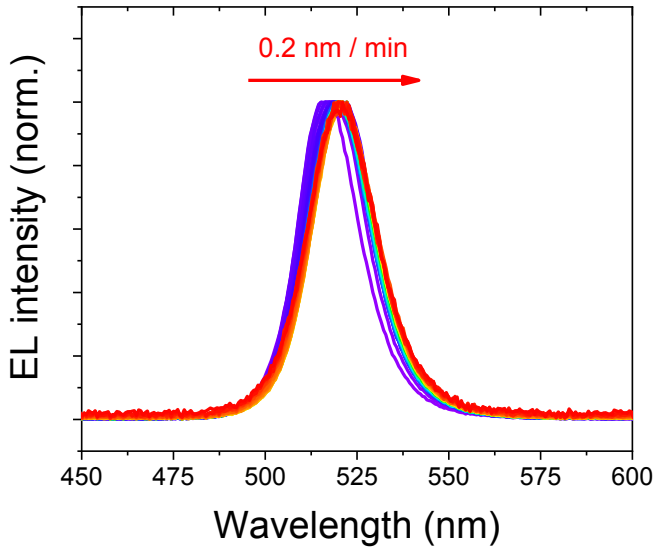

Figure S4. Electroluminescence spectra as a function of time while biasing 0D/3D/0D device ($Cs_4PbCl_6/CsPbBr_3/Cs_4PbCl_6$) at 100 A/m^2 for 20 minutes.

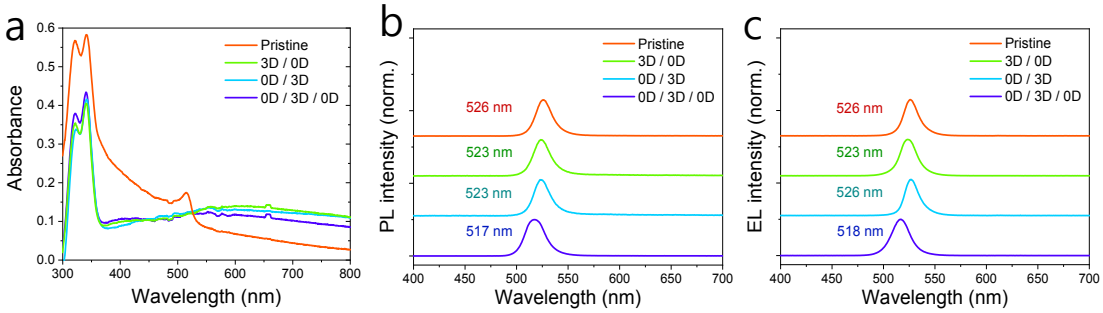

Figure S5. a) Optical absorption, b) photo- and c) electro-luminescence spectra from pristine $CsPbBr_3$:PEO (3D) films, 3D/0D, 0D/3D and 0D/3D/0D bi- and tri-layer perovskite heterojunctions.

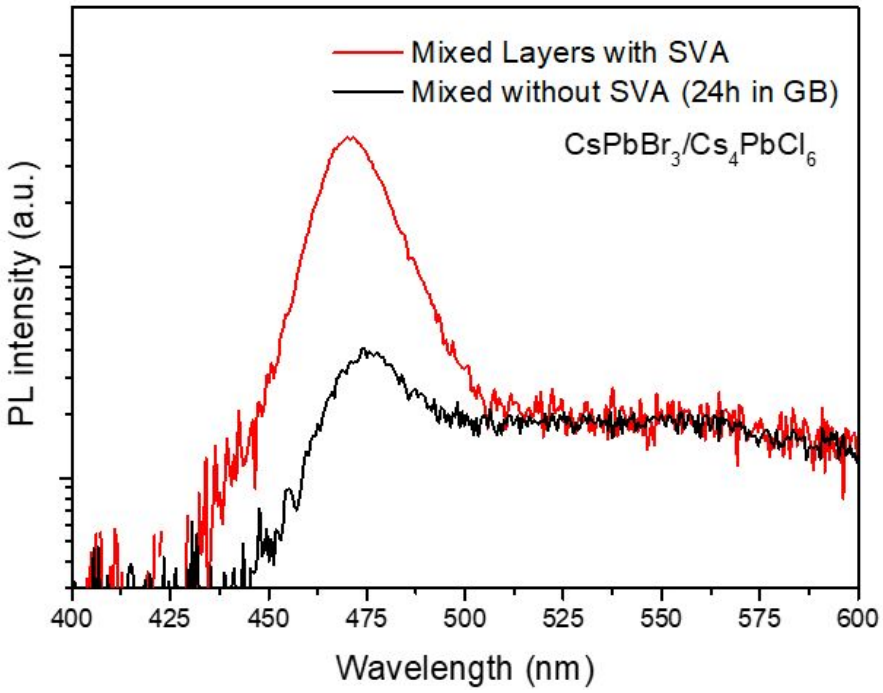

Figure S6. Photoluminescent spectra of intermixed $CsPbBr_3/Cs_4PbCl_6$ with and without solvent vapor annealing (SVA).

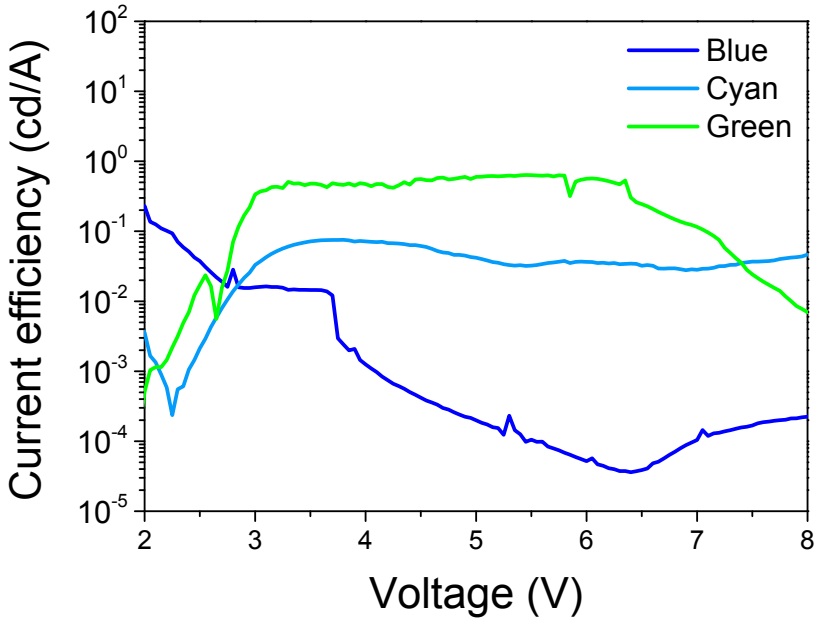

Figure S7. Current efficiency of $CsPbBr_3$ and Mixed $CsPbBr_3/Cs_4PbCl_6$-based devices.

References

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(3) S. Tao, I. Schmidt, G. Brocks, J. Jiang, I. Tranca, K. Meerholz, S. Olthof, *Nat. Commun.* **2019**, *10*, 2560.