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SUPPLEMENTARY INFORMATION

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High-efficiency perovskite–polymer bulk heterostructure light-emitting diodes

Baodan Zhao[1], Sai Bai[2,3], Vincent Kim[1], Robin Lamboll[1], Ravichandran Shivanna[1], Florian Auras[1], Johannes M. Richter[1], Le Yang[1,4], Linjie Dai[1], Mejd Alsari[1], Xiao-Jian She[1], Lusheng Liang[5], Jiangbin Zhang[1], Samuele Lilliu[6,7], Peng Gao[5], Henry J. Snaith[2], Jianpu Wang[8], Neil C. Greenham[1], Richard H. Friend[1*] and Dawei Di[1*]

[1]Cavendish Laboratory, University of Cambridge, Cambridge, UK. [2]Department of Physics, University of Oxford, Clarendon Laboratory, Oxford, UK. [3]Department of Physics, Chemistry and Biology (IFM), Linköping University, Linköping, Sweden. [4]Institute of Materials Research and Engineering (IMRE), Agency for Science, Technology and Research (A*STAR) , Singapore, Singapore. [5]Laboratory of Advanced Functional Materials, Xiamen Institute of Rare Earth Materials, Haixi Institute, Chinese Academy of Sciences, Xiamen, China. [6]Department of Physics and Astronomy, University of Sheffield, Sheffield, UK. [7]The UAE Centre for Crystallography, Abu Dhabi, United Arab Emirates. [8]Institute of Advanced Materials (IAM), Nanjing Tech University, Nanjing, China. *e-mail: rhf10@cam.ac.uk; dd403@cam.ac.uk

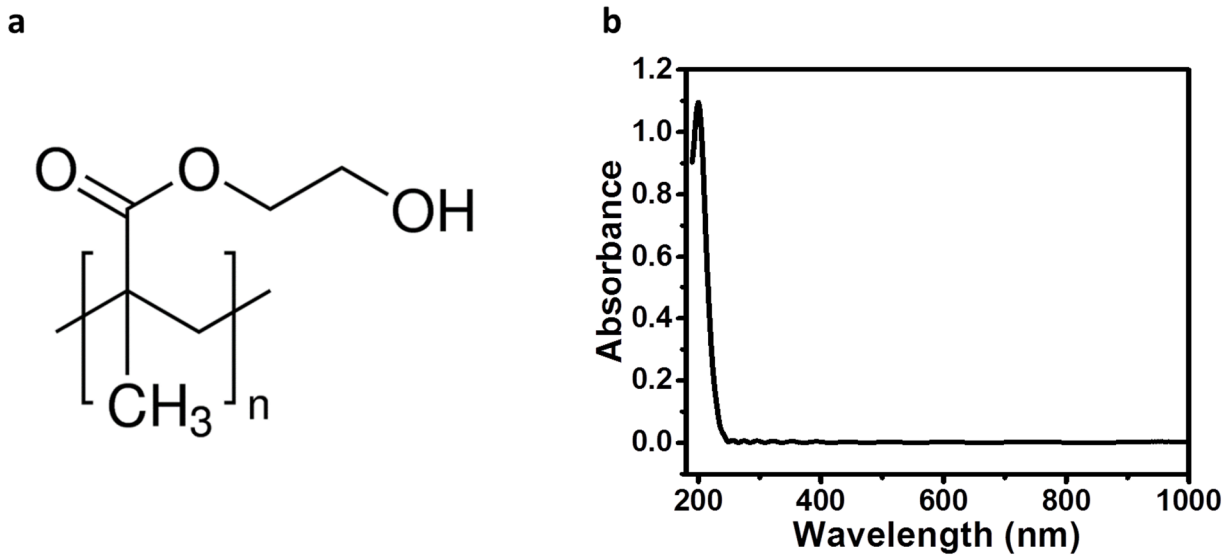

Figure S1 | Further details of poly-HEMA. a, Chemical structure. **b**, UV-Vis absorbance of poly-HEMA, showing an optical bandgap of ~4.96 eV.

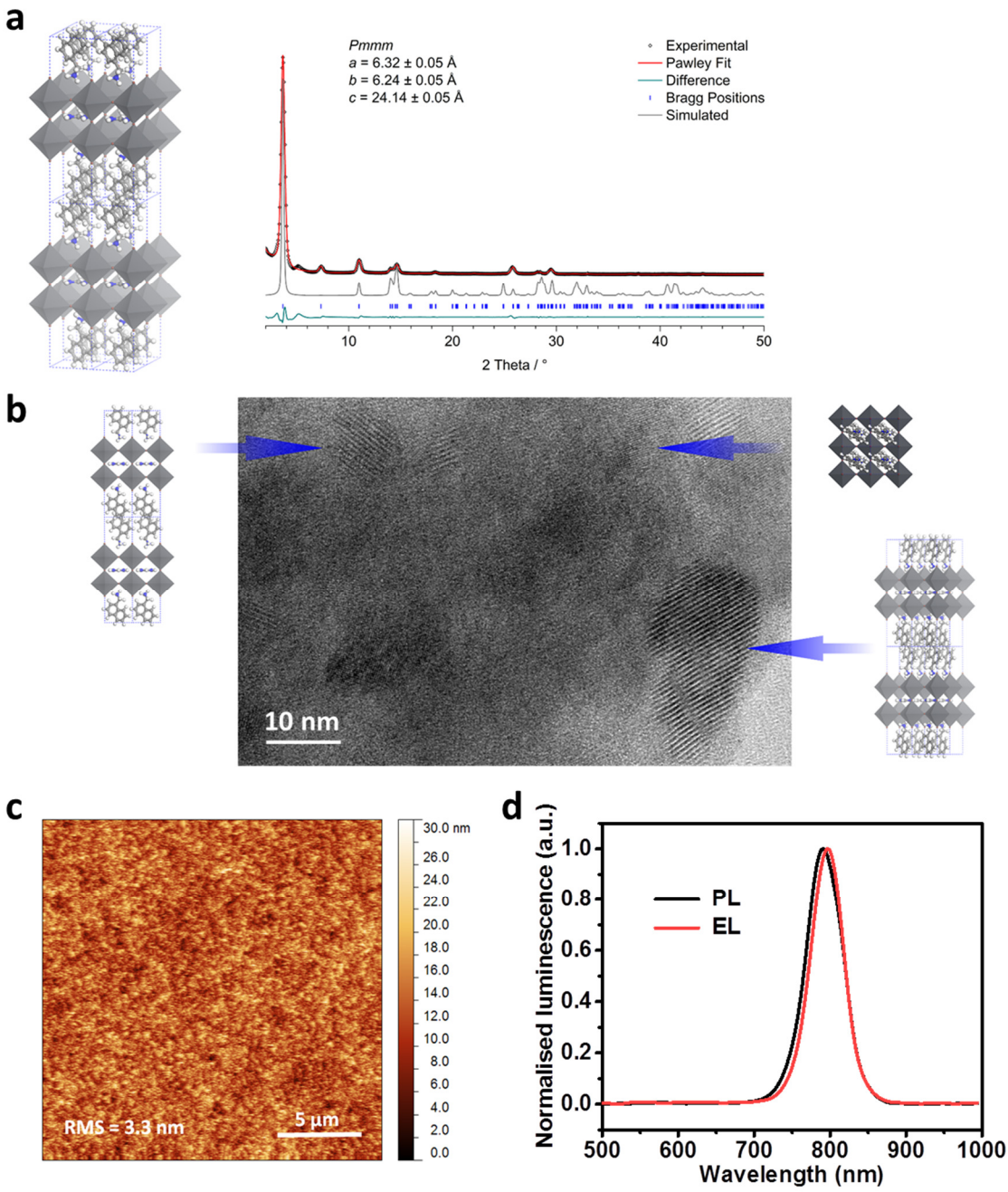

Figure S2 | Further material characterisation of the PPBH film. a, Experimental XRD pattern of the PPBH film (black dots). Pawley refinement (red line) using the force-field optimised structure model shown on the left reproduces the experimental data very well and confirms that the crystalline regions of the films consist almost exclusively of the quasi-2D $(NMA)_2(FA)Pb_2I_7$ phase (grey). For clarity, only one of the respective symmetry-equivalent orientations of the organic cations is displayed. The broadened reflection at 5° might indicate the presence of a small 2D $(NMA)_2PbI_4$ impurity. Scherrer analysis on the reflections at 3.6° and 11.0° reveals average crystallite sizes of 55 nm and 30 nm, respectively. **b**, High-resolution TEM image showing $(NMA)_2(FA)Pb_2I_7$ crystallites in different orientations. **c**, AFM image of the PPBH film, showing an RMS roughness of 3.3 nm. **d**, PL and EL spectra of the PPBH.

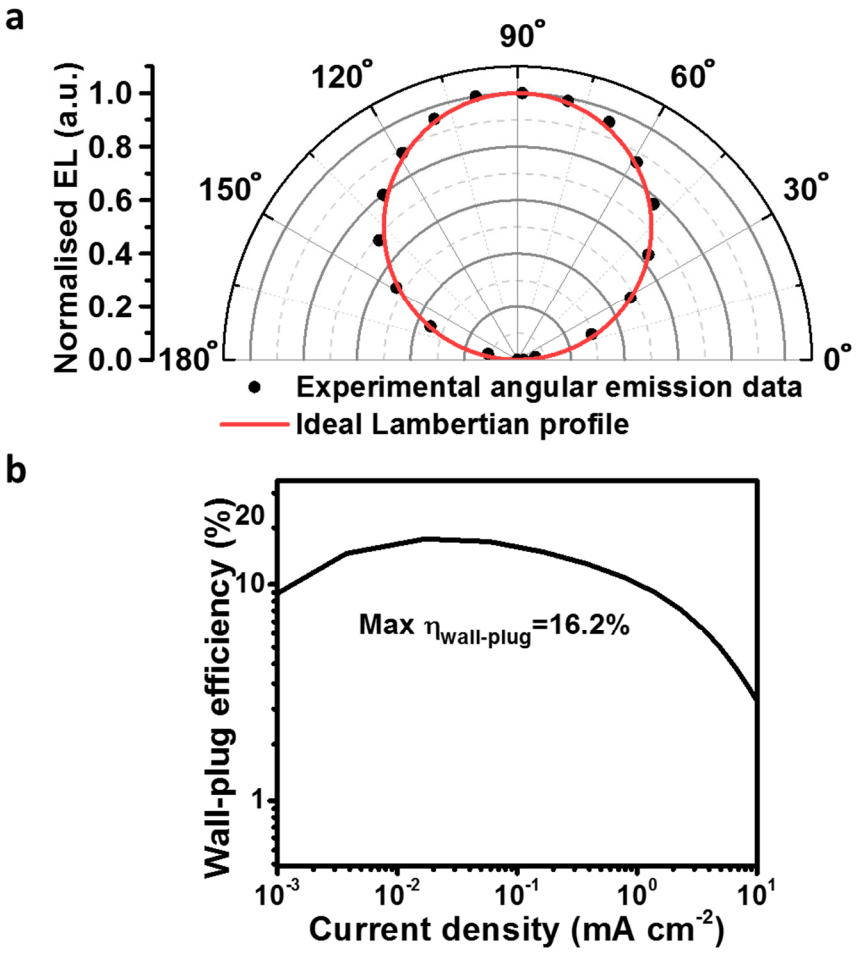

Figure S3 | Additional LED data. a, Angular emission profile of a PPBH LED. **b**, Wall-plug efficiency (electricity-to-light power-conversion efficiency) of the best PPBH LED.

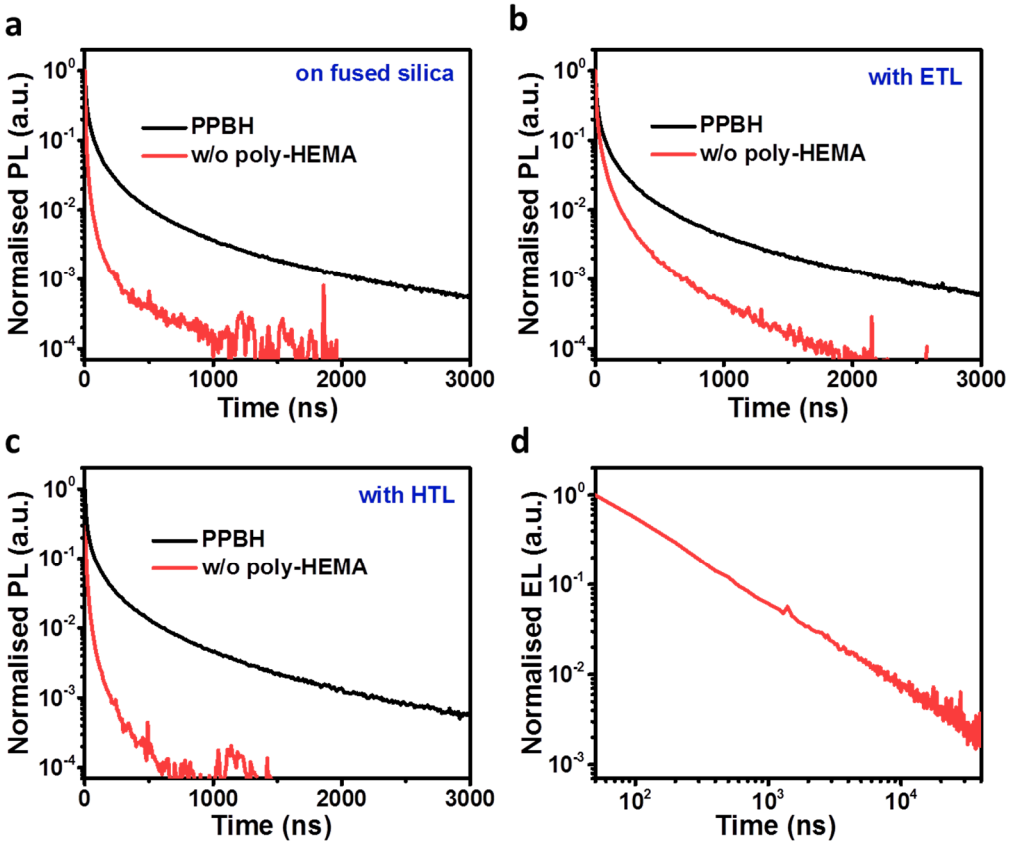

Figure S4 | Charge-transport-interface-dependent transient luminescence data. a-c, PL decay kinetics of PPBH films and identically-prepared quasi-2D/3D perovskite films without the poly-HEMA polymer component in contact with different materials; **a**, on fused silica substrates (without charge-transport layers); **b**, with electron-transport layer (ETL); **c**, with hole-transport layer (HTL). For the PL measurements, the excitation source was a 400-nm, 80-fs pulsed laser with a repetition frequency of 1 kHz. The excitation fluence was 5 μJ cm^{-2}. **d**, Transient EL measurements of a PPBH LED. During the transient EL measurements, the device was driven by 2 V/0 V (on/off) square voltage pulses at a frequency of 1 kHz. The EL decay was recorded during the off-cycles of the pulses, with 50-ns time-steps. It should be noted that the transient EL kinetics can be influenced by other processes such as the measurement circuit RC discharge rate, and the transit time of the slower carriers.

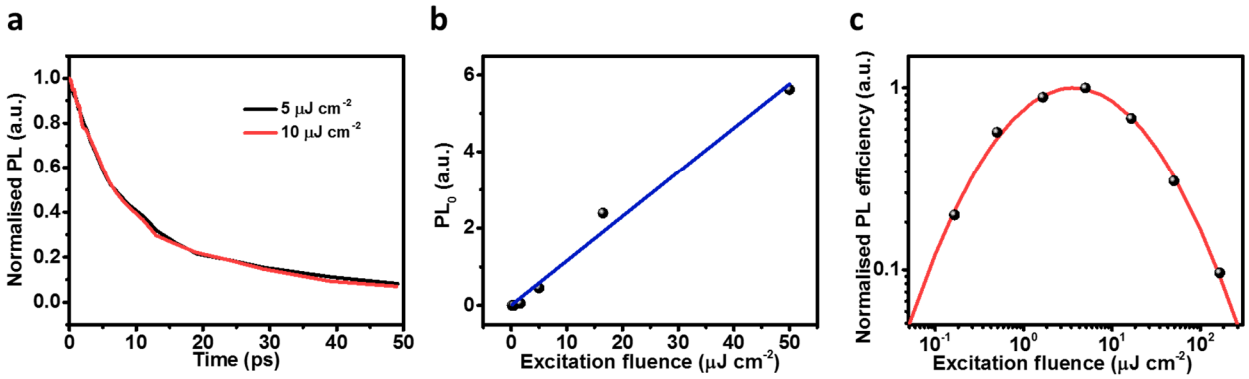

Figure S5 | Further PL data collected using fs laser excitation. a, Ultrafast (fs-ps) PL decay kinetics of the PPBH at 550-620 nm. **b**, Initial PL intensity (PL counts at $t \approx 0$ ns), as a function of excitation fluence. **c**, Fluence-dependent PL efficiency of the PPBH film. For these measurements, the excitation source was a 400-nm, 80-fs pulsed laser with a repetition frequency of 1 kHz.

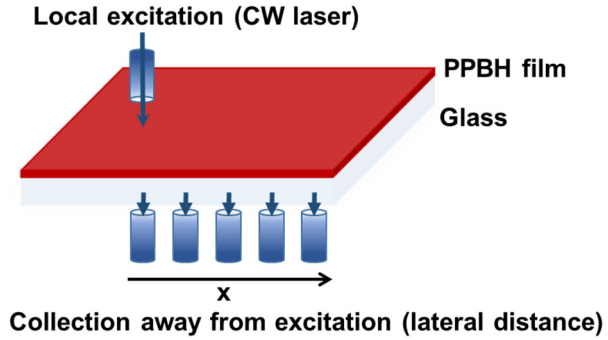

Figure S6 | Lateral PL experimental setup.

Table S1 | Photophysical parameters extracted from various transient optical measurements of the PPBH system. 'Excitation transfer' denotes the ultrafast migration of localised higher-energy excitations from the quasi-2D perovskite $(NMA)_2(FA)Pb_2I_7$ to delocalised lower-energy excitations (charges) on the quasi-3D perovskite sites, characterised using the TA experiments. 'Early-time (fs-ps) PL decay' is the initial PL decay kinetics observed in the quasi-2D perovskite $(NMA)_2(FA)Pb_2I_7$, measured using the fs-ps ultrafast PL setup. 'Bimolecular' denotes the bimolecular (band-to-band) radiative recombination processes in the quasi-3D perovskite domains, characterised using the ns-µs transient PL setup. 'Monomolecular' stands for the first-order (exponential) decay processes observed from the ns-µs PL decay tail kinetics. k and τ represent the rate constant and lifetime of each process, respectively.

Excitation transfer (quasi-2D/3D)		**Early-time (fs-ps) PL decay (quasi-2D)**		**Bimolecular (quasi-3D)**	**Monomolecular (quasi-3D)**	
Transient absorption		Ultrafast (fs-ps) transient PL		ns-µs transient PL	ns-µs transient PL	
k (s^{-1})	τ (s)	k (s^{-1})	τ (s)	k ($cm^3\ s^{-1}$)	k (s^{-1})	τ (s)
1.0×10^{12}	1.0×10^{-12}	1.0×10^{11}	1.0×10^{-11}	3.2×10^{-11}	6.7×10^{5}	1.5×10^{-6}

Table S2 | Parameters for optical modelling.

Layer	Thickness (nm)	Refractive index (~795 nm)
Air	-	1
Au	80	0.124+5.009i
TFB-PFO/MoO_x	57	1.57+0.04i
PPBH	180	1.9
MZO/PEIE	20	1.9
ITO	150	1.61+0.0056i
Glass	1×10^6	1.5
Air	-	1