Supporting Information

Ion Migration-Induced Degradation and Efficiency Roll-off in Quasi-2D Perovskite Light-Emitting Diodes

Tai Cheng,^{†,‡} Ganbaatar Tumen-Ulzii,^{†,‡} Dino Klotz,[§] Satoru Watanabe,^{†,‡} Toshinori Matsushima, *^{‡,§} and Chihaya Adachi*^{†,‡,§}

[†]Center for Organic Photonics and Electronics Research (OPERA), Kyushu University, 744 Motooka, Nishi, Fukuoka 819-0395, Japan

[‡]Japan Science and Technology Agency (JST), ERATO, Adachi Molecular Exciton Engineering Project, 744 Motooka, Nishi, Fukuoka 819-0395, Japan

[§]International Institute for Carbon-Neutral Energy Research (WPI-I2CNER), Kyushu University, 744 Motooka, Nishi, Fukuoka 819-0395, Japan

*E-mail: tmatusim@i2cner.kyushu-u.ac.jp (T.M.).

*E-mail: adachi@cstf.kyushu-u.ac.jp (C.A.).



Figure S1. (a) EQE versus current density curves of a 3D and quasi-2D PeLEDs. (b) Evolutions of the voltage and EQE of an 3D PeLED under constant-current operation. Since the 3D PeLED showed no EL at 0.25 mA cm^{-2} , a constant current of 1 mA cm^{-2} was applied during the measurement.



Figure S2. (a) Voltage and EQE evolutions of an $n_r = 6$ PeLED under constant-current operation with rests in certain operation times. Each rest lasts 10 min. (b) TRPL data (dots) along with fitting curves of an $n_r = 6$ PeLED measured in the start, end, and rest times in Figure S1a. The calculated average PL lifetimes are displayed in the figure.



Figure S3. Discretely collected EQE values of $n_r = 9$ PeLEDs during a rest at the 300 s of the constantcurrent operation. During the rest, a voltage of 0 or -1 V were applied to the devices.



Figure S4. Normalized PL spectra corresponding to the PLQY measurement points in Figure 2e, in logarithmic scale.



Figure S5. (a)-(e) Current kinetics of an $n_r = 9$ PeLED under the driving of the voltage pulses for calculating transient EQE. A rising of current during the pulse can be observed in all the cases as the voltage increased from 5.3 V to 17.6 V, which is corresponding to the voltage dropping during the first decay stage in Figure 1c. Although the trends of current during the pulses in Figure S2e are hard to recognize, they are increasing with slopes that are similar to in Figure S2d.



Figure S6. Current, luminance and EQE kinetics of an $n_r = 9$ PeLED during voltage pulses of (a) 9.6 V and (b) 16.6 V.



Figure S7. Evolutions of (a) current density and (b) EQE of $n_r = 6$ and $n_r = 9$ PeLEDs under the driving of a constant voltage of 4.5 V. The raising and dropping stages of current density correspond to the dropping and raising stages of the voltage evolution under constant-current driving (Figure 1c).



Figure S8. AC impedance spectra of an $n_r = 9$ PeLED before and after the constant-current measurement. The solid curves were fitted with the equivalent circuit as illustrated in the inset. The calculated R_p values are displayed aside of the curves.



Figure S9. (a)-(d) Voltage pulse durations and the corresponding luminance evolutions of an $n_r = 9$ PeLED.



Figure S10. Voltage evolutions of EODs, HODs and perovskite-only devices.



Figure S11. (a) Profile XPS peak intensity curves of $n_r = 9$ PeLEDs before (solid lines) and after (dash lines) degraded to the second stage. (b) Enlarged figure of (a) to show the clear difference of the C 1s curves. The blue arrows point the change directions after the degradation.