Dynamic Redistribution of Mobile Ions in Perovskite Light-Emitting Diodes

Heyong Wang, Zhan Chen, Jingcong Hu, Hongling Yu, Chaoyang Kuang, Jiajun Qin, Xianjie Liu, Yue Lu, Mats Fahlman, Lintao Hou, Xiao-Ke Liu,* and Feng Gao*

Despite quick development of perovskite light-emitting diodes (PeLEDs) during the past few years, the fundamental mechanisms on how ion migration affects device efficiency and stability remain unclear. Here, it is demonstrated that the dynamic redistribution of mobile ions in the emissive layer plays a key role in the performance of PeLEDs and can explain a range of abnormal behaviours commonly observed during the device measurement. The dynamic redistribution of mobile ions changes charge-carrier injection and leads to increased recombination current; at the same time, the ion redistribution also changes charge transport and results in decreased shunt resistance current. As a result, the PeLEDs show hysteresis in external quantum efficiencies (EQEs) and radiance, that is, higher EQEs and radiance during the reverse voltage scan than during the forward scan. In addition, the changes on charge injection and transport induced by the ion redistribution also well explain the rise of the EQE/radiance values under constant driving voltages. The argument is further rationalized by adding extra formamidinium iodide (FAI) into optimized PeLEDs based on FAPbI3, resulting in more significant hysteresis and shorter operational stability of the PeLEDs.

1. Introduction

Perovskite light-emitting diodes (PeLEDs) present great opportunities toward a new generation of low-cost and high-performance LED technology, because of the excellent properties of metal halide perovskites, such as high photoluminescence quantum yields (PLQYs), readily tunable emission spectra, and solution processability.[1] Since the first reported room-temperature PeLEDs in 2014,[2] the external quantum efficiencies (EQEs) of PeLEDs have quickly progressed to over 20% through rational materials and device engineering.[3] However, some phenomena, such as hysteresis in EQE–voltage curves[2] and enhanced EQE under electric fields,[4] cause challenges on reliable characterizations of PeLEDs; lack of understanding of these phenomena also limits further development of high-efficiency and stable PeLEDs.

For perovskite solar cells, ion migration in the perovskite layer is considered as a main reason behind the hysteresis phenomenon and the short operational stability.[5] For PeLEDs, there are also scattered reports about the ion migration.[2,6] The influence of ion migration on PeLEDs is supposed to be more significant than that on perovskite solar cells due to two reasons: (a) PeLEDs operate under much higher bias voltages than perovskite solar cells; (b) excess ions (with a much higher ratio than those in perovskite solar cells) are frequently used for achieving high-efficiency PeLEDs.[1b,d,6a] Therefore, it is highly desired to systematically understand the influence of mobile ions on the performance of PeLEDs.

Here, we study the impact of mobile ions in the emissive layer on the performance of PeLEDs. We observe obvious hysteresis in EQE– and radiance–voltage curves between forward and reverse voltage scans. In addition, obvious rises in EQE/radiance can be observed under constant driving voltages below a critical point, which is found to be around the voltage for achieving the peak EQE. We also find that the dynamic redistribution of mobile ions in the emissive layer is responsible for these phenomena. The redistribution of mobile ions decreases the net electric field in the emissive layer and induces band bending at the interfaces between the emissive layer and charge transport layers, resulting in decreased shunt resistance current and increased recombination current at working voltages, respectively. We further confirm our findings by adding extra
organic salts (e.g., FAI) beyond the optimization ratio, hence introducing more mobile ions and resulting in aggravated hysteresis and decreased operational stability.

2. Results

2.1. The “Rise” Phenomena in PeLEDs

Efficient and stable PeLEDs reported in our previous work[7] are used in this study. Figure 1a illustrates the device architecture, which consists of indium tin oxide (ITO)/polyethyleneimine ethoxylated (PEIE)-modified zinc oxide (PEIE-ZnO, approximately 50 nm)/emissive layer (approximately 70 nm)/poly(9,9-diocetyl-fluorene-co-N-(4-butylphenyl)diphenylamine) (TFB, approximately 60 nm)/molybdenum oxide (MoO<sub>x</sub>, 7 nm)/gold (Au, 60 nm). The emissive layer is a perovskite-molecule composite thin film, consisting of in situ formed high-quality 3D FAPbI<sub>3</sub> nanocrystals embedded in an electron–transport molecular matrix (4,4′-diaminodiphenyl sulfone, DDS).

We find that the PeLED exhibits higher EQEs and radiance in reverse voltage scan (from high to low voltages) than in forward scan (from low to high voltages), and that the difference becomes smaller at higher driving voltages. We perform the voltage scans from 0 V to 3.0 V and then back to 0 V. Note that the highest voltage of 3.0 V in the scans does not induce permanent degradation of the PeLED. Compared with the results during the forward scan, as shown in Figure 1b,c, the device exhibits higher EQE (up to 4.2%) and radiance (up to 3.2 W m<sup>-2</sup> sr<sup>-1</sup>) values during the reverse scan. Meanwhile, as shown in Figure 1d, the relative ratios of enhanced EQE and radiance decrease with increasing driving voltages. As shown in Figure S1, Supporting Information, the same tendency is observed when we switch the scan order, that is, from 2.5 V to 0 V and back to 2.5 V.

We also monitor the EQEs under various constant driving voltages. As shown in Figure 1e, at a constant driving voltage of 1.4 V, the EQEs rise remarkably in the first few seconds and double in values after 15 s. At higher bias voltages (i.e., 1.6 V and 2.0 V), the EQE still increases with increasing time, but much less significantly. Further increasing the driving voltage to 3.0 V, the EQE rise phenomenon disappears; in contrast, the EQE starts decreasing at the initial operating time in this case. The light outputs show a similar tendency at various constant driving voltages (Figure S2, Supporting Information). Hence, we believe that there is a critical voltage, below which the device shows the rises in EQE and light output during the initial operating time. Moreover, we notice that the critical voltage is located around the voltage for reaching peak EQE (around 2.0 V for this PeLED as shown in Figure 1b).

2.2. Dynamic Redistribution of Mobile Ions

We speculate that the above-mentioned phenomena are associated with the mobile ions in the emissive layer. Besides the
intrinsic ionic species in the perovskite crystals, the excess FAI may remain in the perovskite film,\(^\text{[8]}\) which can also result in mobile ions. In addition, we experimentally find excess iodide ions on the surface of the perovskite nanocrystals for our samples. As shown in Figure S3a, Supporting Information, the XRD pattern of the perovskite film shows pure 3D FAPbI\(_3\) crystal structure. The absorption and PL spectra (Figure S3b, Supporting Information) also show features similar to those of the 3D FAPbI\(_3\) film.\(^\text{[7]}\) These results indicate that the resulting perovskite film is pure 3D FAPbI\(_3\) without other perovskite phases. However, as shown in Figure 2a, the X-ray photo-electron spectroscopy (XPS) result shows that the I signal of this film is much stronger than its Pb signal under the same measurement conditions. Calculated by the fitted areas of the XPS peaks and the relative atomic sensitivity factor, the molar ratio between I and Pb is 6.2 to 1.0, indicating that the perovskite film contains excess iodide ions. To confirm this result, we carry out scanning transmission electron microscopy (STEM) and energy-dispersive X-ray spectroscopy (EDS) measurements. As shown in the cross-sectional high-angle annular dark field (HAADF) STEM image (Figure 2b) and associated EDS line scan (Figure 2c) of the device, the surface of the perovskite particle has a higher I/Pb ratio than the interior. These results indicate that excess iodide ions exist on the surface of the perovskite particles, which might be the source of mobile ions during device operation.\(^\text{[9]}\)

In order to better explain the impact of the mobile ions on the device performance, we start with a simple situation where the amount of generated charge carriers is fixed by optically exciting the sample with a laser (450 nm), and focus on the driving voltages in the range before the injection occurs (in Figure 3a). This is roughly the situation of a photovoltaic device at voltages below the open-circuit voltage. As a result, the electric fields in the emissive layer of the PeLED are similar to those in perovskite solar cells, consisting of an external electric field \(E_{\text{ex}}\) generated by a driving voltage, a built-in electric field \(E_{\text{bi}}\) resulting from the device structure, and an ion redistribution associated electric field \(E_{\text{red}}\).\(^\text{[10]}\) These three electric fields add up to a net electric field \(E_{\text{net}}\).

The net electric field determines the recombination, extraction, and injection of charge carriers. The generated photocarriers can be either extracted out of the device (resulting in photocurrent) or recombine in the emissive layer (the radiative recombination part results in photoluminescence, PL). Since electroluminescence (EL) is detectable when the driving voltages are larger than 0.98 V mainly in the dark condition (Figure S4, Supporting Information), the luminescence detected below 0.98 V is mainly dominated by PL while that above 0.98 V is a mixture of PL and EL. As shown in Figure 3b, the PL intensity increases with increasing driving voltage from −0.2 to 0.98 V. This can be explained by the reduced net electric field due to the increased external electric field in the emissive layer, suppressing the extraction of photogenerated charge carriers and facilitating the recombination in the emissive layer.\(^\text{[11]}\)

The presence and redistribution of mobile ions change the net electric field, resulting in obvious hysteresis in the PL intensity- and current–voltage curves between forward and reverse voltage scans. Compared with the PL intensities during the forward scan, the device exhibits lower PL intensities in the reverse scan in a driving voltage region of 0–0.98 V. We can exclude the possibility that this observation is caused by trap filling process, which can reduce the trap density of the perovskite and result in enhanced PL intensity.\(^\text{[12]}\) We can also exclude the possibility of the perovskite degradation causing the PL decrease in the reverse scan (as we discuss in the next paragraph). Instead, this observation (decreased PL intensities during the reverse scan) is well explained by the increased net electric field, which results from the redistribution of mobile ions, as discussed in detail in Note S1, Supporting Information. This explanation is also consistent with the hysteresis of the current–voltage curve, which shows higher values in the reverse scan. Similar hysteresis of the current–voltage curves has been widely observed in perovskite solar cells and is attributed to ion migration.\(^\text{[5]}\) We further calculate the voltage variations \(\Delta V_{\text{in}}\left(\Delta V_{\text{in}} = V_{\text{reverse}} - V_{\text{forward}}\right)\), where \(V_{\text{reverse}}\) and \(V_{\text{forward}}\) are the driving voltages at the same PL intensity during the reverse and forward scans, respectively) caused by the redistribution of mobile ions in the emissive layer; as shown in Figure 3c, \(\Delta V_{\text{in}}\) is in the range of 0.02–0.12 V during the voltage scan from 0–0.98 V.

With good understanding of the steady-state process from the discussions above, we now move forward to further understand the dynamic redistribution progress of the mobile ions by performing quasi steady-state measurements. We measure the changes of the PL intensity and photocurrent upon the application of a constant external driving voltage of 0.8 V. As shown in Figure 3d, the PL intensity is constant without the

---

**Figure 2.** a) XPS spectra of I 3d and Pb 4f peaks of the perovskite film. b) Cross-sectional HAADF-STEM image and c) the related EDS profiles of I and Pb elements from the line scan of the PeLED.
external bias, indicating that the degradation in the emissive layer is negligible during the measurement. Upon the application of a bias of 0.8 V, the PL intensity immediately drops to around 70% of its initial value, which then shows two slow rise processes (with time constants of 1.2 and 14.4 s), as shown in Table S1, Supporting Information. In the meanwhile, the photocurrent intensity shows two slow decay processes with similar time constants as the PL case. The slow enhancement of the PL intensity and the slow decay of the photocurrent indicate that the net electric field continuously yet slowly decreases as a result of the redistribution of mobile ions, in spite of a constant external electric field. In addition to this situation where we apply a bias to a device without any bias pre-treatment, our model can also well explain the other situations where the device is pre-treated by an external bias before the measurement, as shown in Figure S5, Supporting Information and discussed in Note S2, Supporting Information.

2.3. The Impact of Mobile Ions on the Performance of PeLEDs

After understanding how the ion redistribution affects the physical processes under illumination, we now demonstrate that ion redistribution also significantly affects the electrical properties of the perovskite in the dark condition (without illumination)—the condition closely related with LED operations. As shown in Figure S6a, Supporting Information, the current flowing through the emissive layer includes shunt resistance current ($I_S$) and recombination current ($I_{Rec}$, which results from the recombination of carriers).

We first find that the redistribution of mobile ions decreases the shunt resistance current which is related to the net electric field. As shown in Figure S6b, Supporting Information, the accumulated mobile ions cause a new electric field, which has a reverse direction to the external electric field and decreases the net electric field in the emissive layer. Under small biases (e.g., 0.8 V as shown in Figure 4a), the current is mainly shunt resistance current. The device exhibits a slow decay of the current density at a constant driving voltage of 0.8 V (Figure S7a, Supporting Information). This decay process is consistent with the reduced net electric field by ion redistribution. As expected, the decay process at 0.8 V is suppressed at lower temperatures (Figure S7b, Supporting Information). The low temperature can suppress ion migration, which reduces the change of net electric field in the emissive layer. In addition, the curve (Figure S7a, Supporting Information) shows a slow decay of negative current density when the driving voltage is switched from 0.8 V to 0 V, indicating a recovery process of the accumulated mobile ions.

Under large biases where the charge injection is obvious (and the device is turned on), the redistribution of mobile ions also helps to increase the recombination current by decreasing the injection barriers for carriers (Figure S6b, Supporting Information). Figure 4b shows the current densities of our PeLED at various driving voltages. As discussed in the previous paragraph, the slow decay of current densities at a driving voltage of 0.8 V is the result of reduced

---

*Figure 3. a) Diagram of the carrier behaviors under electric fields, where $E_{ex}$, $E_{bi}$, $E_{in}$, and $E_n$ are external electric field, built-in electric field, ions redistribution associated electric field, and net electric field, respectively. b) Current density–voltage–luminescence intensity curves in the forward and reverse voltage scans. c) Mobile ion-induced voltage variations $\Delta V_{in}$ in the emissive layer. d) Normalized photocurrent and PL intensity curves under illumination without or with a constant driving voltage of 0.8 V.*
shunt resistance current. However, increasing the driving voltages to 1.0 and 1.1 V, where the device exhibits injection character as shown in Figure 4a, the PeLED shows a slightly increased current density after a drop in the first several seconds. Under these driving voltages, the accumulated mobile ions at the interfaces between the emissive layer and the transport layers induce band bending, which decreases the injection barriers between the emissive layer and the transport layers and hence increases the recombination current density. Hence, this phenomenon is caused by the combined impacts of the decreased shunt resistance current and the increased recombination current. Further increasing the driving voltage to high values (e.g., 1.2, 1.6, and 2.0 V), a fast rise of the current density is observed without the decay process during our measurement, indicating that the shunt resistance current is negligible compared with the recombination current at high driving voltages. At a driving voltage of 3.0 V, the device shows continuously increased current density, possibly due to the permanent degradation of the device during this continuous measurement.

With all these understandings of how the ion redistribution affects the electrical properties of PeLEDs under different biases, we can now rationalize the abnormal phenomena observed in Figure 1. The reduced shunt resistance current improves the injection efficiency and is more significant in the low driving voltages where the shunt resistance current has a large ratio in the current, resulting in the obviously enhanced EQE at low driving voltages. Meanwhile, the increased current density shifts the efficiency to higher value before reaching the peak EQE (around 2.0 V in our PeLED). However, further increasing the driving voltage (e.g., 3.0 V) beyond the peak EQE point, the increased current density shifts the efficiency to lower value, resulting in a quickly dropped EQE. As such, the hysteresis and the "rise" phenomena in Figure 1 can be well explained.

2.4. Modulation of Mobile Ions in PeLEDs

In order to further validate our discussions, we design a new experiment by adding extra mobile ions based on the optimal blend and investigating how the extra mobile ions affect our device properties. We prepare two perovskite films from the perovskite precursor solutions with different amounts of excess FAI (FAI/PbI₂ = 2.2 and 2.5 in molar ratio, where 2.2 is the optimal ratio in our previous work(7)). As shown in Figure S8a, Supporting Information, the absorption and PL spectra of the films (FAI/PbI₂ = 2.2 and 2.5) are almost the same, attributed to 3D FAPbI₃ perovskite. Moreover, as shown in Figure S8b, Supporting Information, the films of FAI/PbI₂ = 2.2 and 2.5 show almost the same PLQY values even under various excitation powers, indicating that the perovskite crystals in these two samples have almost the same quality.

The devices based on the resulting perovskite films exhibit decreased current densities and decreased radiances with increasing amount of excess FAI (Figure 5a), indicating the decreased conductivity of the perovskite film caused by excess FAI. The normalized EL spectra of these two devices are identical, peaking at 802 nm (Figure 5b), which is consistent with the PL results (Figure S8a, Supporting Information). Moreover, as shown in Figure 5c, the FAI/PbI₂ = 2.5 based device delivers slightly higher peak EQE than the FAI/PbI₂ = 2.2 based device; meanwhile, the former operates at a much lower current density than the latter to reach the peak EQE. However, as shown in Figure 5d and Table S2, Supporting Information, the FAI/PbI₂ = 2.5 based device exhibits worse operational stability.

We further perform the forward and reverse voltage scans between 0–3 V and calculate the EQEs at each voltage. As shown in Figure 5e, both devices show hysteresis of EQE–voltage curves. The FAI/PbI₂ = 2.5 based device shows aggravated hysteresis (Figure 5f), which can be ascribed to the extra mobile ions caused by excess FAI in this device. These results agree well with our discussions above and suggest that rational modulation of the excess ions in the perovskite emissive layer is desired to reduce the detrimental effects of the mobile ions and to achieve PeLEDs with both high efficiency and long operational stability.

3. Conclusions

In summary, we have observed the hysteresis in the EQE and radiance curves with higher EQEs and radiances during the reverse voltage scan than those obtained during the forward scan. Moreover, the EQE and radiance rise at constant driving voltages below a critical point, which is around the voltage for the peak EQE. We have also found that these phenomena can
be well–explained by the dynamic redistribution of mobile ions in the emissive layer. The redistribution of the mobile ions decreases shunt resistance current and increases recombination current, resulting in the “rise” phenomena. In addition, by introducing extra FAI in the emissive layer, we have demonstrated that excess mobile ions in the device can aggravate the hysteresis in the EQE–voltage curve and shorten the operational stability of PeLEDs. This work provides new understanding of the redistribution of mobile ions in PeLEDs and their impact on the device performance, which will inspire further development of PeLEDs with both high efficiency and long operational stability.

4. Experimental Section

Materials: Colloidal ZnO nanocrystals were synthesized following the reported solution-precipitation process.[7] Formamidinium iodide (FAI) was obtained from Dyesol, and PbI₂ was obtained from TCL (≥ 98.0% purity). Other chemicals were obtained from Sigma-Aldrich.

Perovskite Precursor Solution Preparation: The perovskite precursor solutions were prepared by dissolving FAI, PbI₂, and 4,4′-diaminodiphenyl sulfone (DDS) in N,N-dimethylformamide (DMF, anhydrous, 99.8%) with a various molar ratio between FAI, PbI₂, and DDS of x: 1: 1 (x = 2.2 or 2.5). The concentration of Pb²⁺ was kept at 0.1 m. The perovskite precursor solutions were put on a hot plate and stirred at 60 °C overnight.

Film Preparation: ITO-coated substrates were cleaned using ultrasonic bath in deionized water for 15 min and then dried by high-speed nitrogen flow, followed by ultraviolet-ozone for 10 min. Colloidal ZnO nanocrystals were deposited on ITO at 4000 rpm for 30 s in air. Then polyethyleneimine ethoxylated (PEIE) in isopropyl alcohol (0.03 wt%) was spin-coated onto the ZnO layer at 5000 rpm for 30 s and then annealed at 100 °C for 5 min. The perovskite precursor solutions were spin-coated onto the PEIE-ZnO films at 4000 rpm for 30 s, followed by thermal annealing at 100 °C for 5 min.

Device Fabrication and Characterization: Based on the film preparation procedures, PeLEDs were completed with the following processes: TFB in chlorobenzene (12 mg mL⁻¹) was spin-coated at 3000 rpm for 30 s. Then the films were transferred to a vacuum chamber (1×10⁻⁶ Torr), in which MoO₃ and Au were deposited at the rates of 0.2 Å s⁻¹ and 1.5 Å s⁻¹, respectively. Characterization of the PeLEDs was carried out at room temperature in a nitrogen (N₂)-filled glovebox. Current density–voltage (J–V) characteristics were recorded by the Keithley 2400 source meter. Forward-viewing spectral radiant flux was measured by an integrating sphere coupled with a QE65 Pro spectrometer. The active device area was 0.0725 cm². The operational stability of the devices (without encapsulation) was measured in a testing box (stored in the N₂-filled glovebox).

Simultaneous Measurement of PL and Photocurrent: The simultaneous measurement of PL intensity and photocurrent were carried out on a homemade setup consisting of a Keithley 2400 and a QE65 Pro spectrometer. A weak continuous-wave 450 nm laser was used as the light source; the light spot was focused within the emitting area of the device.

Film Characterization: X-ray diffraction patterns were obtained from an X-ray diffractometer (Pananalytical X’Pert Pro) with an X-ray tube (Cu Kα, λ = 1.5406 Å). Steady-state PL spectra of the films were recorded by using a 450 nm laser as an excitation source. Absorption spectra were measured with a PerkinElmer model Lambda 900. XPS tests were carried out by using a Scienta ESCA 200 spectrometer in ultrahigh vacuum (approximately 1×10⁻¹⁰ mbar) with a monochromatic Al (K alpha) X-ray source providing photons with 1486.6 eV. TEM specimens were prepared using a focused ion beam (FEI Helios NanoLab 600i) operating at 2–30 kV. Cross-sectional HAADF-STEM and line scan EDS were carried out with an FEI Titan G2 60–300 microscope at 300 kV.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.
Acknowledgements

The authors thank Y. Wang, H. Zhang, C. Bao, W. Tress, and W.-X. Ni for helpful discussions. This work was supported by the ERC Starting Grant (No. 717026), the Swedish Energy Agency Energimyndigheten (No. 48758-1 and 44651-1), the Swedish Foundation for International Cooperation in Research and Higher Education (No. CH2018-7736), and the Swedish Government Strategic Research Area in Materials Science on Functional Materials at Linköping University (Faculty Grant SFO-Mat-LiU No. 2009-00971). L.H. thanks the NSFC Project (61774077) and the Research and Development Program in Key Areas of Guangdong Province (2019B1515120073, 2019B09021002, 2019B01032004) for financial support. H.W., H.Y., and C.K. acknowledge the financial support of the China Scholarship Council. F.G. is a Wallenberg Academy Fellow.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

efficiency, LEDs, mobile ions, perovskite, stability

Received: September 5, 2020
Revised: October 31, 2020
Published online: November 23, 2020

[1] a) M. Sessolo, L. Gil-Escrig, G. Longo, H. J. Bolink, Top. Curr. Chem. 2016, 374, 52; b) Q. Van Le, H. W. Jang, S. Y. Kim, Small Methods 2018, 2, 1700419; c) X. Zhao, J. D. A. Ng, R. H. Friend, Z.-K. Tan, ACS Photonics 2018, 5, 3866; d) M. Lu, Y. Zhang, S. Wang, J. Guo, W. W. Yu, A. L. Rogach, Adv. Funct. Mater. 2019, 29, 1902008; e) X. K. Liu, W. Xu, S. Bai, Y. Jin, J. Wang, R. H. Friend, F. Gao, Nat. Mater. 2020, https://doi.org/10.1038/s41563-020-0784-7.

[2] Z.-K. Tan, R. S. Moghaddam, M. L. Lai, P. Docampo, R. Higler, F. Deschler, M. Price, A. Sadhanala, L. M. Pazos, D. Credgington, F. Hanusch, T. Bein, H. J. Snaith, R. H. Friend, Nat. Nanotechnol. 2014, 9, 687.

[3] a) Y. Cao, N. Wang, H. Tian, J. Guo, Y. Wei, H. Chen, Y. Miao, W. Zou, K. Pan, Y. He, H. Cao, Y. Ke, M. Xu, Y. Wang, M. Yang, K. Du, Z. Fu, D. Kong, D. Dai, Y. Jin, G. Li, H. Li, Q. Peng, J. Wang, W. Huang, Nature 2018, 562, 249; b) W. Xu, Q. Hu, S. Bai, C. Bao, Y. Miao, Z. Yuan, T. Borzda, A. J. Barker, E. Tyukalova, Z. Hu, M. Kawecki, H. Wang, Z. Yan, X. Liu, X. Shi, K. Uvdal, M. Fahlman, W. Zhang, M. Duchamp, J.-M. Liu, A. Petrozza, J. Wang, L.-M. Liu, W. Huang, F. Gao, Nat. Photonics 2019, 13, 418; c) T. Chiba, Y. Hayashi, H. Ebe, K. Hoshi, J. Sato, S. Sato, Y.-J. Pu, S. Ohisa, J. Kido, Nat. Photonics 2018, 12, 681; d) K. Lin, J. Xing, L. N. Quan, F. P. G. de Arquer, X. Gong, J. Lu, L. Xie, W. Zhao, D. Zhang, C. Yan, W. Li, X. Liu, Y. Lu, J. Kirman, E. H. Sargent, Q. Xiong, Z. Wei, Nature 2018, 562, 245; e) B. Zhao, S. Bai, V. Kim, R. Lamboll, R. Shivanova, F. Auras, J. M. Richter, L. Yang, L. Dai, M. Alsari, X.-J. She, L. Liang, J. Zhang, S. Lilliu, P. Gao, H. J. Snaith, J. Wang, N. C. Greenham, R. H. Friend, D. Di, Nat. Photonics 2018, 12, 783.

[4] a) L. Zhao, J. Gao, Y. L. Lin, Y. W. Yeh, K. M. Lee, N. Yao, Y. L. Loo, B. P. Rand, Adv. Mater. 2017, 29, 1605317; b) W. Zou, R. Li, S. Zhang, N. L. Wang, Y. Cao, Y. Miao, M. Xu, Q. Guo, D. Di, L. Zhang, C. Yi, F. Gao, R. H. Friend, J. Wang, W. Huang, Nat. Commun. 2018, 9, 608.

[5] H. J. Snaith, A. Abate, J. M. Ball, G. E. Eperon, T. Leijtens, N. K. Noel, S. D. Stranks, J. T. Wang, K. Wójcickowski, W. Zhang, J. Phys. Chem. Lett. 2014, 5, 1511.

[6] a) Z. Xiao, R. A. Kerner, L. Zhao, N. L. Tran, K. M. Lee, T.-W. Koh, G. D. Scholes, B. P. Rand, Nat. Photonics 2017, 11, 108; b) Y. Zhou, H. Xue, Y. H. Jia, G. Brooks, S. Tao, N. Zhao, Adv. Funct. Mater. 2019, 29, 1905739.

[7] H. Wang, F. U. Kosasih, H. Yu, C. Zheng, J. Zhang, G. Pozina, Y. Liu, C. Bao, Z. Hu, X. Liu, L. Kobera, S. Abbrert, J. Brus, Y. Jin, M. Fahlman, R. H. Friend, C. Ducati, X. K. Liu, F. Gao, Nat. Commun. 2020, 11, 891.

[8] Y. H. Jia, S. Neutzner, Y. Zhou, M. Yang, J. M. F. Tapia, N. Li, H. Yu, J. Cao, J. P. Wang, A. Petrozza, C. P. Wong, N. Zhao, Adv. Funct. Mater. 2019, 30, 1906875.

[9] a) H. Jin, E. Debroye, M. Keshavarz, I. G. Schebykin, M. B. J. Roelfers, J. Hofkens, J. A. Steele, Mater. Horiz. 2019, 7, 397; b) Y. Yuan, J. Huang, Acc. Chem. Res. 2016, 49, 286.

[10] Z. Xu, T. De Rosia, K. Weeks, J. Phys. Chem. C 2017, 121, 24389.

[11] T. Leijtens, A. R. Srimath Kandada, G. E. Eperon, G. Grancini, V. D’Innocenzo, J. M. Ball, S. D. Stranks, H. J. Snaith, A. Petrozza, J. Am. Chem. Soc. 2015, 137, 15451.

[12] D. W. deQuilettes, W. Zhang, V. M. Burlakov, D. J. Graham, T. Leijtens, A. OsheroV, V. Bulovic, H. J. Snaith, D. S. Ginger, S. D. Stranks, Nat. Commun. 2016, 7, 11683.

[13] B. Chen, M. Yang, X. Zheng, C. Wu, W. Li, Y. Yan, J. Bisquert, G. Garcia-Belmonte, K. Zhu, S. Priya, J. Phys. Chem. Lett. 2015, 6, 4693.

[14] D. Kruangam, M. Deguchi, T. Toyama, H. Okamoto, Y. Hamakawa, IEEE Trans. Electron Devices 1988, 35, 957.

[15] G.-B. Lin, D. Meyaard, J. Cho, E. F. Schubert, H. Shim, C. Sone, Appl Phys Lett. 2012, 100, 161106.

[16] J. Wang, N. Wang, Y. Jin, J. Si, Z. K. Tan, H. Du, L. Cheng, X. Dai, S. Bai, H. He, Z. Ye, M. L. Lai, R. H. Friend, W. Huang, Adv Mater. 2015, 27, 2311.