







Fig. 1 Schematic of the n-i-p PSC architecture employing (a) c-SnO<sub>2</sub>, (b) c-NP-SnO<sub>2</sub>, (c) c(Li)-SnO<sub>2</sub>, and (d) c(Li)-NP-SnO<sub>2</sub> ETLs. Statistical photovoltaic parameters for 15 PSCs each employing the various ETL configurations in reverse scan direction: (e) PCE, (f) FF, (g) V<sub>oc</sub>, (h) J<sub>sc</sub>, (i) Statistical hysteresis index (HI) extracted from J-V curves.

charge carrier extraction during the first seconds of device operation, as recently reported by Thiesbrummel *et al.*<sup>66</sup> We also fabricated PSCs with a single SnO<sub>2</sub> NP layer (NP-SnO<sub>2</sub>) as it is a well-established ETL in the literature<sup>15-18,31,55,63,67,68</sup> and our

laboratory.<sup>69-71</sup> The champion PSC with NP-SnO<sub>2</sub> ETL shows a reverse scan PCE and s-PCE of 19.3% and 16.3% respectively (Fig. S7, ESI<sup>†</sup>), which is much lower compared to the values achieved for the champion c(Li)-NP-SnO<sub>2</sub> ETL (Fig. 2d). Besides,



Fig. 2 Reverse (R) and forward (F) J-V scans and stabilized PCE (s-PCE) derived from MPP tracking of champion PSCs with (a and e) c-SnO<sub>2</sub>, (b and f) c-NP-SnO<sub>2</sub>, (c and g) c(Li)-SnO<sub>2</sub> and (d and h) c(Li)-NP-SnO<sub>2</sub> ETLs.



the PSCs with c(Li)-NP-SnO<sub>2</sub> ETL maintain a stable power output (>95% of the initial value) when measured under 1 Sun illumination for 24 h (Fig. S8, ESI†).

To identify the reasons for the differences in performance of PSCs employing the various ETL configurations, we firstly probe the ETL surface morphology by means of atomic force microscopy (AFM) and scanning electron microscopy (SEM). The AFM images depict that the morphology for the c-SnO<sub>2</sub> ETL (Fig. 3a) appears quite similar to that of ITO (Fig. S9a, ESI†), while the RMS roughness is slightly reduced from 2.9 nm to 2.2 nm. This observation is attributed to the existence of a thin and continuous SnO<sub>2</sub> compact layer on ITO. The c(Li)-SnO<sub>2</sub> ETL also exhibits a similar morphology and RMS roughness (2.1 nm) as compared the c-SnO<sub>2</sub> ETL without any observable aggregation of Li-TFSA salt (Fig. 3b). After deposition of SnO<sub>2</sub> NPs on top of the c-SnO<sub>2</sub> or c(Li)-SnO<sub>2</sub> layers, the film morphology slightly changes accompanied by a reduced RMS surface roughness of 1.3 nm and 1.4 nm, respectively, which is attributed to the formation of a smooth NP capping layer that homogeneously covers the existing bumps of the underlying layers.<sup>17,72</sup>

Comparable morphology and RMS are also observed when SnO<sub>2</sub> NPs are deposited directly on bare ITO (Fig. S9b, ESI†). In addition, the SEM images demonstrate a similar surface morphology for the different ETL configurations in line with the results obtained by AFM (Fig. S10a–d, ESI†). The above observations are also in line with the comparable transmittance and reflectance spectra for the different ETLs (Fig. S11a and b, ESI†).

Furthermore, the transmission in the wavelength region ~450 nm is only slightly reduced as compared to bare ITO, resulting in a very low parasitic absorption of the respective PSCs.

Next, we examine if employing the different ETL configurations does alter the perovskite thin film morphology, thickness and crystallinity. SEM images in Fig. 3e–l and XRD spectra in Fig. S12 (ESI†) indicate that all perovskite thin films on the different ETLs exhibit a pin-hole free morphology with comparable grain size, thickness and XRD pattern. These findings confirm that the ETL sublayer has no significant effect on the perovskite film formation. The zoom-in of the cross-sectional SEM images of the of the ETLs/perovskite thin film interface indicates a similar thickness for the c-SnO<sub>2</sub> and c(Li)-SnO<sub>2</sub> ETLs, while that for c-NP-SnO<sub>2</sub> and c(Li)-NP-SnO<sub>2</sub> ETLs is slightly increased (Fig. 3i–l inset, highlighted in green). We note that we do not expect the c-SnO<sub>2</sub> layer (annealed at 200 °C) to be washed away or be redissolved by the water-based NP-SnO<sub>2</sub> deposition, in line with previous reports.<sup>63</sup>

Next, we investigate the surface chemistry of the various ETL configurations by means of X-ray photoelectron spectroscopy (XPS) to proof the presence of Li and/or K at the film surface and to study any potential effect on the chemical environment of SnO<sub>2</sub>. Fig. 4a shows the survey spectra for the full accessible binding energy range, where the core level emissions were identified from reference values (see dotted lines and labels).<sup>73</sup> All four ETLs exhibit the expected lines related to tin (Sn) and oxygen (O). Furthermore, carbon (C) is present because

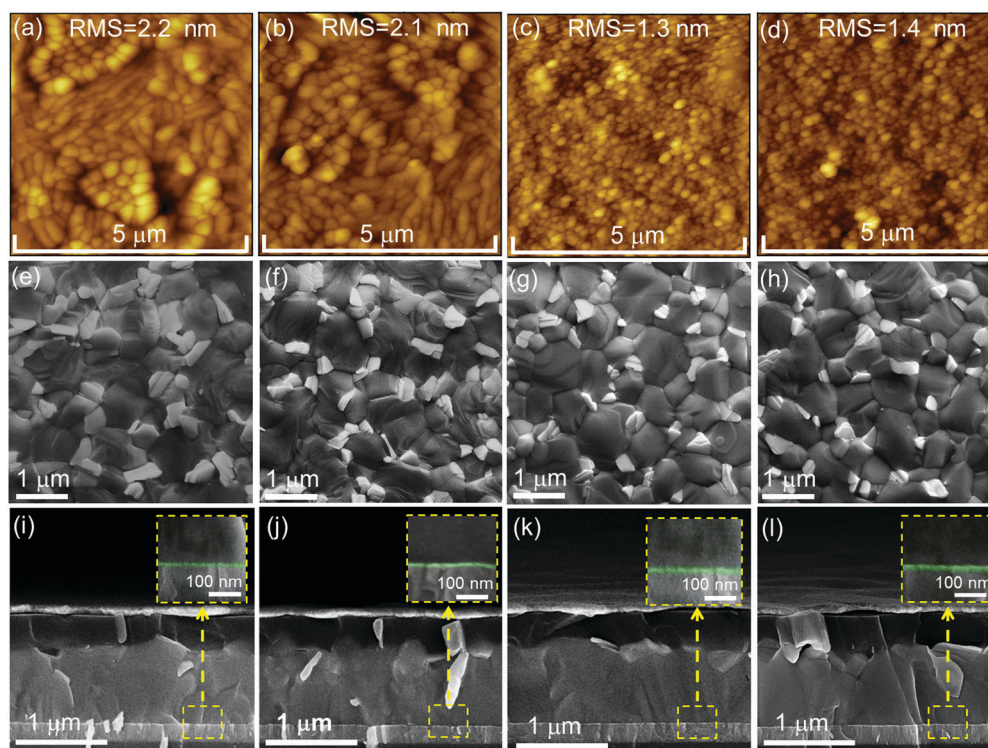


Fig. 3 AFM images of the surface morphology of (a) c-SnO<sub>2</sub>, (b) c(Li)-SnO<sub>2</sub> (c) c-NP-SnO<sub>2</sub> and (d) c(Li)-NP-SnO<sub>2</sub> ETLs. Top and cross-sectional SEM images of perovskite thin films deposited on (e and i) c-SnO<sub>2</sub>, (f and j) c(Li)-SnO<sub>2</sub>, (g and k) c-NP-SnO<sub>2</sub> and (h and l) c(Li)-NP-SnO<sub>2</sub> ETLs.





Fig. 4 (a) Survey spectra, (b) Li 1s emission, and (c) Sn 3d<sub>5/2</sub> emission of the four different ETL configurations c-SnO<sub>2</sub>, c-NP-SnO<sub>2</sub>, c(Li)-SnO<sub>2</sub>, and c(Li)-NP-SnO<sub>2</sub> measured by XPS. For shape comparison, the spectra in (c) were normalized and shifted.

of the *ex situ* preparation and sample transfer and handling in ambient air. For c-SnO<sub>2</sub> and c(Li)-SnO<sub>2</sub>, a Cl 2p signal around 200 eV is detected, which is attributed to residuals of the SnCl<sub>2</sub> precursor (see Experimental section). In the case of c-NP-SnO<sub>2</sub> and c(Li)-NP-SnO<sub>2</sub>, small potassium (K) peaks at ~294 eV (K 2p<sub>3/2</sub>) and ~297 eV (K 2p<sub>1/2</sub>) are detected (see Fig. S13, ESI†), which is attributed to residuals from KOH that is used for the synthesis of the NPs to stabilize the colloids as discussed above.<sup>63</sup>

Despite lithium having a relatively low photoionization cross-section,<sup>74</sup> *i.e.* it is barely detectable in the XPS survey spectrum, it can be unambiguously identified for the c(Li)-SnO<sub>2</sub> ETL from the Li 1s core level spectra at a binding energy of ~56 eV (Fig. 4b). This indicates that Li ions have been successfully doped into SnO<sub>2</sub>. In addition, small peaks related to the fluorine F 1s and sulfur S 2p core levels at ~689 eV and ~170 eV originating from TFSI<sup>-</sup> anions at the film surface are detected in the survey spectrum (see Fig. 4a), in line with recent observations for Li-TFSI doped mesoporous TiO<sub>2</sub>.<sup>5</sup> Critically, the signals of F, S, and Li disappear for the c(Li)-NP-SnO<sub>2</sub> ETL as their signal gets damped by the NPs. This reveals that there is no diffusion of the dopants from c(Li)-SnO<sub>2</sub> through the SnO<sub>2</sub> NPs towards the surface as otherwise the signal from the diffused dopant would be no longer be damped by the NPs and be still visible in the spectrum. The absence of a Li signal thereby shows that the Li-TFSI treatment affects only the interface between c-SnO<sub>2</sub> and NP-SnO<sub>2</sub>. In Fig. 4c, we compare the normalized and shifted Sn 3d<sub>5/2</sub> emission spectra for all different ETLs, which do not show any variations in the shape of the signal, indicating that the chemical environment of SnO<sub>2</sub> is the same for all four ETL configurations. We note, however,

that the Sn 3d<sub>5/2</sub> emission is not very sensitive to chemical shifts;<sup>75,76</sup> thus, an influence of the Li-TFSI treatment might not be detectable. The comparable morphological and structural properties of the different ETLs and perovskite thin films on top mean that the difference in the photovoltaic performance must originate from changes in the optoelectronic properties at the ETL/perovskite interface. Therefore, we investigate the recombination behaviour and charge transfer kinetics between the perovskite and the different ETLs by means of photoluminescence quantum efficiency ( $Q_e^{\text{lum}}$ ) and time resolved photoluminescence (TRPL).

The normalized steady state PL spectra of perovskite films deposited on the various ETL configurations measured in an integrating sphere exhibit a similar PL peak shape (Fig. S14, ESI†). This indicates that the perovskite bandgap and morphology are not strongly affected by the underlying ETL,<sup>77</sup> in line with the absorbance, SEM, AFM and XRD results. In order to determine the quasi-Fermi level splitting ( $q \cdot \Delta E_F = V_{\text{OC,rad}} + k_B T \ln Q_e^{\text{lum}}$ ), we measure  $Q_e^{\text{lum}}$  of perovskite films deposited on ITO/ETL substrates at irradiation intensities comparable to 1 Sun illumination as a measure for the maximum obtainable  $V_{\text{OC}}$  (*i.e.*, the implied  $V_{\text{OC,imp}} = \frac{\Delta E_F}{q}$ ) when employing this interface in a PSC.<sup>78–80</sup>

As shown in Fig. 5a, a  $Q_e^{\text{lum}}$  of 5% (measured directly after turning on the laser) for perovskite films on a glass substrate relates to  $V_{\text{OC,imp}}$  of ~1.13 V, while when deposited on the different ETL configurations  $Q_e^{\text{lum}}$  slightly reduces to ~1–2% which relates to  $V_{\text{OC,imp}}$  in the range of ~1.1–1.12 V. The extracted values are comparable to the average (reverse scan)  $V_{\text{OC}}$  of the respective PSCs in the  $J$ - $V$  characteristics (see Fig. 2), indicating that the  $V_{\text{OC}}$  is limited by the ETL/perovskite interface. We note that while the perovskite/HTL interface can also limit the  $V_{\text{OC}}$  substantially in certain device structures, as has been shown in several recent reports,<sup>67,69,70,81,82</sup> this interface is not the focus of this study and does not seem to result in a large additional interface recombination for our PSCs. Based on these results we can conclude that a reduction in interface recombination cannot explain the superior properties (enhanced  $J_{\text{SC}}$  and FF and lower hysteresis) of PSCs employing the c(Li)-NP-SnO<sub>2</sub> ETL, which is in line with the comparable reverse scan  $V_{\text{OC}}$  for the different ETLs.<sup>82</sup> For this reason, we measured the TRPL decay for ITO/ETL/perovskite stacks to further analyze the charge extraction and non-radiative recombination properties (Fig. 5b). Despite the complicated interpretation of such transients,<sup>83–86</sup> at early times (*i.e.*, high initial charge carrier densities) the decay is affected by charge transfer to the ETL. At later times (*i.e.*, at low charge carrier densities), the decay is mainly governed by Shockley–Read–Hall (SRH) recombination at either the ETL/perovskite interface or within the bulk of the perovskite thin film and can be approximately described by a constant exponential slope.<sup>79,85,86</sup> Strikingly, the decay at early times (<100 ns) is much faster for the c(Li)-NP-SnO<sub>2</sub> ETL as compared to the other ETL configurations that all exhibit a very similar decay, indicating towards improved electron extraction capabilities of this specific ETL configuration.<sup>15,85–87</sup> One reason for that observation might be a lower amount of charge accumulation at this interface, which





**Fig. 5** (a) Left y-axis: External photoluminescence quantum efficiency ( $Q_e^{\text{lum}}$ ) of perovskite films on glass and on ITO/ETL substrates for the different ETLs. Right y-axis: Implied  $V_{\text{OC}}$  calculated either via  $V_{\text{OC,imp}} = V_{\text{OC,rad}} + \frac{kT}{q} \cdot \ln Q_e^{\text{lum}}$  or  $V_{\text{OC,imp}} = \frac{kT}{q} \cdot \ln \frac{G_{\text{ext}} \Delta \tau_{\text{SRH}}}{n_0}$ , (b) time resolved photoluminescence (TRPL) measurements of perovskite films on the various ITO/ETL configurations, (c) time dependent measurements of  $Q_e^{\text{lum}}$  at two different irradiation intensities of perovskite films on the various ITO/ETL configurations.

otherwise would quickly slow down the charge extraction process at early times.<sup>83,85,86</sup> A single exponential fit to the data at later times ( $\sim 300$  ns) allows estimating the SRH lifetime ( $\tau_{\text{SRH}}$ ) which is very similar for c-SnO<sub>2</sub>, c-NP-SnO<sub>2</sub> and c(Li)-SnO<sub>2</sub> ETLs

( $\tau_{\text{SRH}} \sim 220$  ns), while it is slightly lower for the c(Li)-NP-SnO<sub>2</sub> ETL ( $\tau_{\text{SRH}} \sim 150$  ns). Assuming that  $\tau_{\text{SRH}}$  is governed by recombination in the bulk of the perovskite films allows estimating  $V_{\text{OC,imp}} = \frac{kT}{q} \ln \frac{G_{\text{ext}} \Delta \tau_{\text{SRH}}}{n_0}$ , where  $G_{\text{ext}}$  is the charge carrier generation rate and  $n_0$  is the thermal equilibrium concentration of electrons.<sup>79,88</sup>

The results of this analysis (average of 4 different spots) are included in Fig. 5a. While the values of  $V_{\text{OC,imp}}$  for the c-SnO<sub>2</sub>, c-NP-SnO<sub>2</sub> and c(Li)-NP-SnO<sub>2</sub> ETLs are slightly below that extracted from  $Q_e^{\text{lum}}$ , that for the c(Li)-SnO<sub>2</sub> ETL is slightly higher, which could indicate that the TRPL decay at later times is governed by additional processes for this ETL.

To explore the effect of ion migration and its effect on hysteresis for the various ETL configurations, we performed continuous light-soaking experiments at low ( $\sim 0.1$  Sun) and high ( $\sim 3$  Sun) irradiation intensity. This allows us to study the photo-brightening effect, which can be used to estimate the magnitude of ion migration and defect healing in a perovskite film (Fig. 5c).<sup>56,89–93</sup> In line with the measurements at 1 Sun intensity discussed above, the initial value of  $Q_e^{\text{lum}}$  at  $\sim 0.1$  Sun is very similar ( $\sim 0.5\%$ ) for all ETL configurations. However,  $Q_e^{\text{lum}}$  for c-SnO<sub>2</sub> as ETL exhibits fast rise dynamics even at these low irradiation intensities, while in contrast the photo-brightening effect is strongly suppressed in case of c(Li)-SnO<sub>2</sub>, and almost completely suppressed for c-NP-SnO<sub>2</sub> and c(Li)-NP-SnO<sub>2</sub>. Increasing the irradiation intensity to  $\sim 3$  Sun where ion migration is expected to be strongly enhanced,<sup>94,95</sup> samples with c-NP-SnO<sub>2</sub>, c(Li)-NP-SnO<sub>2</sub> and c(Li)-SnO<sub>2</sub> as ETL exhibit similar rise dynamics, while that for c-SnO<sub>2</sub> is again much stronger. Importantly, returning to the low intensity of  $\sim 0.1$  Sun, c-NP-SnO<sub>2</sub>, c(Li)-SnO<sub>2</sub> and c(Li)-NP-SnO<sub>2</sub> ETLs all exhibit a  $Q_e^{\text{lum}}$  that is around a factor  $\sim 1.6$  higher compared to the value just before the high intensity step, while, in contrast,  $Q_e^{\text{lum}}$  for c-SnO<sub>2</sub> as ETL remains at a very high value of 5%, which is a factor  $\sim 2.5$  higher. This reduced photo-brightening effect and suppressed hysteresis of  $Q_e^{\text{lum}}$ , when going from low-to-high-to-low intensities, indicates that ion migration is strongly suppressed when employing c-NP-SnO<sub>2</sub> and c(Li)-NP-SnO<sub>2</sub> as ETL and to some extent also for c(Li)-SnO<sub>2</sub>.<sup>56,68,89,90,93</sup>

In summary, the  $Q_e^{\text{lum}}$  and TRPL results reveal that the degree of interface recombination at the ETL/perovskite interface is not strongly affected by the different ETL configurations, while there is an indication for a more efficient electron extraction in case of c(Li)-NP-SnO<sub>2</sub>. In addition, ion migration seems to be strongly reduced for samples employing c-NP-SnO<sub>2</sub> and c(Li)-NP-SnO<sub>2</sub> as ETL with respect to the reference c-SnO<sub>2</sub> ETL. We hypothesize that the suppressed ion migration is related to a reduced charge accumulation at the ETL/perovskite interface and hence allows faster charge extraction, in line with the strongly reduced hysteresis in the respective PSCs.<sup>92</sup> It should be noted that from the results so far it is not completely clear why the hysteresis in c(Li)-SnO<sub>2</sub> is much higher as compared to c-NP-SnO<sub>2</sub> and c(Li)-NP-SnO<sub>2</sub>.

Therefore, to further explore and understand the charge transport dynamics in PSCs with the various ETL configurations,



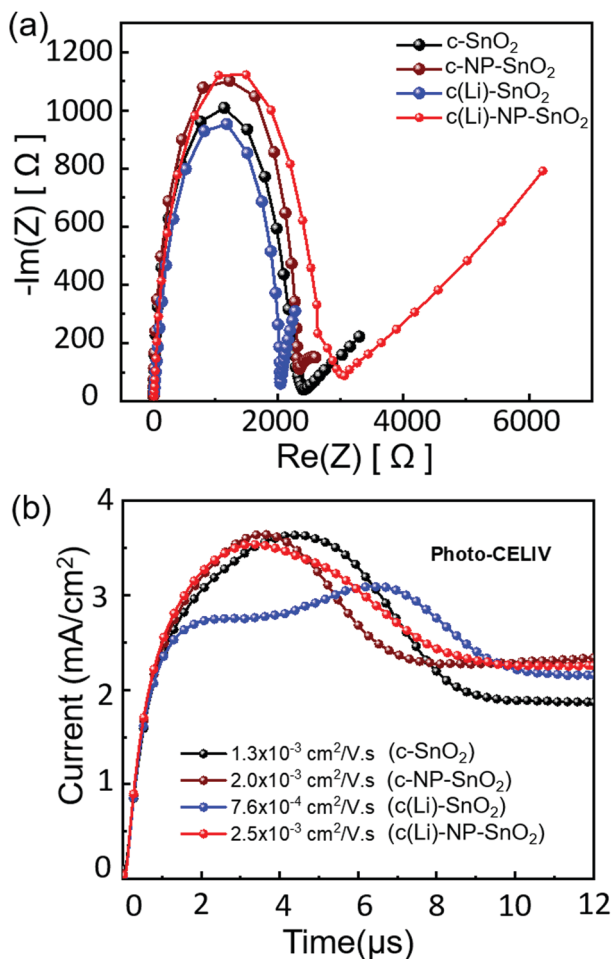


Fig. 6 (a) Electrochemical impedance spectroscopy (EIS) and (b) photo-generated charge extraction by linearly increasing voltage (photo-CELIV) measurements of PSCs with  $\text{c-SnO}_2$ ,  $\text{c(Li)-NP-SnO}_2$  and  $\text{c(Li)-NP-SnO}_2$  ETLs.

we conduct electrochemical impedance spectroscopy (EIS) measurements to characterize the recombination behavior. The EIS measurements are performed under open-circuit conditions at  $3 \text{ mW cm}^{-2}$  intensity. The Nyquist plots of representative PSCs with the different ETL configurations feature two kinetically separated processes with clearly distinguishable semi-circles, recorded from 1 Hz to 1 MHz frequency (see Fig. 6a). As reported by Garcia-Belmonte *et al.* and others,<sup>96–98</sup> the semi-circle in the high frequency range (left side) represents the dielectric properties of the bulk of the perovskite film, and the semi-circle at low frequency range (right side) correlates to the recombination at the interface of the perovskite thin film and its charge transport layers. The impedance response of PSCs is interpreted in terms of the equivalent circuit shown in Fig. S15 (ESI<sup>†</sup>) where  $R_s$  is the series resistance and  $C_b$  and  $R_1$  refer to the capacitance and transport resistance of the bulk perovskite thin film, respectively. The chemical capacitance  $C_\mu$  correlates to the charge accumulation at the interfaces of the perovskite and transport layers and the recombination resistance ( $R_{\text{rec}}$ ) refers to the recombination resistance at the

interfaces.<sup>33,99</sup> The fitted parameters are summarized in Table S1 (ESI<sup>†</sup>). We observe that  $R_{\text{rec}}$  is highest in PSCs with  $\text{c(Li)-NP-SnO}_2$  as ETL followed by  $\text{c-NP-SnO}_2$ ,  $\text{c-SnO}_2$  and  $\text{c(Li)-SnO}_2$ , reflecting the lowest recombination in PSCs with  $\text{c(Li)-NP-SnO}_2$  as ETL compared to the others. Since the HTL remains identical for all PSCs, the change in  $R_{\text{rec}}$  is supposed to be originated from the ETL/perovskite interface. The value of  $C_\mu$  suggests that charge accumulation at the ETL/perovskite interface decreases in the order  $\text{c(Li)-SnO}_2 > \text{c-SnO}_2 > \text{c-NP-SnO}_2 > \text{c(Li)-NP-SnO}_2$ . As mentioned previously, charge accumulation at the perovskite interfaces has been demonstrated to be one of the dominant reasons for  $J-V$  hysteresis.<sup>20–22</sup> The observed trend in charge accumulation behavior is consistent with the TRPL results and the fact that PSCs with  $\text{c(Li)-NP-SnO}_2$  and  $\text{c-NP-SnO}_2$  ETLs show the lowest hysteresis (Fig. 2d), while PSCs with  $\text{c-SnO}_2$  and  $\text{c(Li)-SnO}_2$  ETL show intermediate and maximum hysteresis, respectively (Fig. 2c and a). We note that this charge accumulation does not seem to affect the non-radiative recombination at the ETL/perovskite interface, as demonstrated by the similar value of  $Q_e^{\text{lum}}$ , and therefore the  $V_{\text{OC}}$ , but does affect the charge extraction properties and thereby the FF,  $J_{\text{SC}}$  and hysteresis.

Finally, to investigate whether charge transport throughout the perovskite absorber layer is also affected by the different ETL configurations and whether it correlates with the trend in device performance, we performed photogenerated charge extraction by linearly increasing voltage (photo-CELIV) measurements. Fig. 6b shows the typical photo-CELIV curves for all the PSCs with the peak position correlating to the mobility of the carriers (see details in the ESI<sup>†</sup>). This is because a peak position at later times indicates that a higher voltage is required for most efficient charge extraction.<sup>100,101</sup> Hence, for  $\text{c(Li)-NP-SnO}_2$  and  $\text{c-NP-SnO}_2$ , comparatively lower voltages compared to  $\text{c-SnO}_2$  and  $\text{c(Li)-SnO}_2$  are required to extract all charge carriers, which we attribute to an enhanced charge accumulation in the latter, in line with the EIS results. In addition, an irregular double peak shape can be observed for  $\text{c(Li)-SnO}_2$  ETL, with the total extracted charge carriers, as derived by integrating over the peak area in Fig. 6b, being strongly reduced compared to the other ETL configurations. The calculated mobilities for  $\text{c(Li)-NP-SnO}_2$ ,  $\text{c-NP-SnO}_2$ ,  $\text{c-SnO}_2$  and  $\text{c(Li)-SnO}_2$  are  $2.5 \times 10^{-3}$ ,  $2.0 \times 10^{-3}$ ,  $1.3 \times 10^{-3}$  and  $7.6 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ , respectively. Therefore, we have strong indication that the ETL configuration not only affects the charge extraction characteristics at the interface, but also improves the mobility within the perovskite film for PSCs with  $\text{c(Li)-NP-SnO}_2$  as ETL as compared to  $\text{c-SnO}_2$  and  $\text{c(Li)-SnO}_2$ .<sup>102–105</sup>

## Conclusions

This work systematically investigates various single- and bilayer  $\text{SnO}_2$  ETLs and the role of dopants and additives therein for use in highly-efficient planar n-i-p PSCs. SEM, XRD, and AFM results show that the different ETL configurations do not affect the perovskite film morphology and crystallinity. XPS data reveals that Li is successfully doped in the  $\text{c(Li)-SnO}_2$  ETL and



that K is apparent in the ETLs formed using a commercial SnO<sub>2</sub> NPs colloidal solution. We introduce a novel bilayer c(Li)-NP-SnO<sub>2</sub> ETL that combines the advantages of Li and K, resulting in an improved charge extraction in conjunction with suppressed ion migration and reduced charge accumulation at the perovskite/ETL interface. The champion double-cation PSC with c(Li)-NP-SnO<sub>2</sub> ETL exhibits a remarkable PCE of 20.4% in the *J*-*V* scan and a stabilized PCE of around 18.5% after MPP tracking for 300 s. This is a result of a strongly reduced hysteresis and improvements in both FF and *J*<sub>SC</sub> as compared to the other optimized ETL configurations. In summary, this work reports on an effective interface engineering approach for perovskite photovoltaics to improve their photovoltaic parameters.

## Author contributions

A. A. E. and M. M. B. share equal contribution for conceiving the idea, developing the ETL optimization and processing of the perovskite solar cells. P. F. performed the photoluminescence quantum yield measurements and analysis. M. R. K. conducted the EIS and photo-CELIV characterizations and analysis, I. M. H. performed the TRPL measurements, which were analysed by P. F. M. F. performed the XPS measurements and analysis. S. T. and T. A. conducted the AFM and SEM measurement, respectively. J. A. S. developed the two-step perovskite deposition recipe. M. M. B., P. F. and U. W. P. drafted the manuscript. All co-authors discussed the paper and revised the manuscript. T. M., J. P. H., B. S. R., U. L. and, U. W. P. were involved in designing the experiments and supervised the work.

## Conflicts of interest

The authors declare no conflict of interest.

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