Supplementary Information for

Tin(IV) Dopant Removal through Anti-Solvent Engineering Enabling Tin Based Perovskite Solar Cells with High Charge Carrier Mobilities

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(SI-1) Device Fabrication and Characterization Methods

1.1 Material Preparation

Lead(II) iodide (PbI₂, 99.99%) was purchased from Tokyo Chemical Industry Co., Ltd. (TCI, Japan). Formamidinium iodide (FAI, \geq 98%), methylammonium iodide (MAI, \geq 98%), caesium iodide (CsI₂), tin fluoride (SnF₂, 99%), tin iodide (SnI₂, 99.99%), and aluminium-doped zinc oxide (AI-ZnO nanoparticle ink, 2.5 wt%) were purchased from Sigma Aldrich (UK). [6,6]-Phenyl-C61-butyric acid methyl ester (PC₆₀BM) was purchased from Solenne B.V. PEDOT:PSS (Hereaus Celvios P VP AI 4083) was purchased from Hereaus. Solvents dimethyl sulfoxide (DMSO, anhydrous, \geq 99.9%), N,Ndimethylformamide (DMF, anhydrous, 99.8%), methanol, chlorobenzene (anhydrous, 99.8%), 2propanol (anhydrous, 99.5%) and toluene (anhydrous, 99.8%) were purchased from Sigma Aldrich (UK). All materials were used without further purification.

1.1.1. Pb-Sn mixed perovskite precursor solution: PbI_2 (TCI, Japan), FAI, MAI, SnF_2 and SnI_2 powders were dissolved in a glass vial with DMF:DMSO 4:1 (v/v) solvent mixture. The final concentrations of the dissolved salts were 0.65 M (0.05 M PbI₂ in excess), 1 M, 0.2 M, 0.12 M and 0.6 M respectively. This solution was stirred continuously for at least 1 h. To this, 42 µl of CsI in DMSO (390 mg/ml) was added and stirred for another 30 min prior to fabrication of devices.

1.1.2. PEDOT:PSS hole transport material: PEDOT:PSS (was filtered through a filter of 0.45 μ m pore size and diluted 1:2 (v/v) in methanol at room temperature.

1.1.3. PC₆₀BM electron transport material: 20 mg of [6,6]-Phenyl-C60-butyric acid methyl ester (PC₆₀BM; Solenne BV) was dissolved in cholobenzene and stirred overnight at room temperature.

1.1.4. ZnO electron transport material: Al doped ZnO nanoparticle ink was diluted 1:1 (v/v) in 2-propanol and sonicated in an ultrasonic bath for 15 min at 100 W at room temperature.

1.2 Devices and Sample Fabrication

1.2.1. Fabrication of Pb-Sn mixed inverted PSC devices: Devices were fabricated on 15 mm x 15 mm x 0.7 mm ITO coated glass substrates (Luminescence Technology Corporation, 15 Ω/\Box), which were first run through a cleaning cycle of sequential sonication in acetone, isopropanol and methanol for 5 min each, in an ultrasonic bath at 100 W, and were subsequently dried using an N₂ gas gun and exposed to O₂ plasma for 5 min to form a hydrophilic surface. The hole transporting layer, PEDOT:PSS solution was then spin coated at room temperature on the ITO substrates, at 5000 rpm, 40 s in air. The substrates were then transferred inside a N₂ glove box and annealed at 150°C for 10 min on a hot plate. Each substrate was then spin coated with 30 µl of perovskite absorber layer sequentially at 1000 rpm for 8 s, and 6000 rpm 37 s. 100 µl of the anti-solvent was dropped 5 s prior to the end of spinning cycle, and the coated substrates were annealed at 70°C for 20 min inside the glove box. The substrates were then brought to room temperature, and each was spin coated with 30µl PC₆₀BM solution as the first electron transport layer, at 1500 rpm, 35 s. As the next electron transport layer, Al doped ZnO nanoparticle solution was then spin coated at 6000 rpm, 30 s. Finally the devices were loaded on a shadow mask and a 7 nm thick bathocuporine (BCP) layer and a 120 nm thick Silver cathode were thermally evaporated under a base pressure of less than 3 x 10⁶ mbar at a rate of 0.3 Å/s.

1.2.2. Sample preparation for UV-Vis spectroscopy: Samples were prepared on cleaned and O_2 plasma treated glass substrates, by spin coating 30 µl of the perovskite solution using the same spinning and anti-solvent dropping parameters as used in solar cell device fabrication. As casted layers were annealed at 70°C for 20 min.

1.2.3. Sample preparation for SEM: Samples were prepared on cleaned and O_2 plasma treated ITO coated glass substrates following the same procedure as in section 1.2.2. Prior to SEM measurements, the edges of the perovskite layers were scratched in order to expose the ITO layer and the sample was grounded using conductive silver paint between the metal SEM stub and the ITO.

1.2.4. Sample preparation for Optical Pump- Terahertz Probe (OPTP) measurements: Samples were prepared on 2mm thick quartz substrates cleaned using sequential sonication in soap water, acetone, IPA and methanol, dried by N_2 gas gun and O_2 plasma treated. 20 µl of the perovskite solutions

was deposited by spin coating using the same parameters as section 1.2.1. The perovskite layers were annealed at 70°C for 20 min.

1.2.5. Sample preparation for GIWAXS measurements: Samples were prepared on Quartz coated glass substrates (20 mm x 15 mm x 1.1 mm, Ossila) cleaned prior to coating of perovskite thin layers. The perovskite coating was prepared inside the glove box following the same spin coating parameters as section 1.2.1)

1.2.6. Sample preparation for X-ray Photoelectron Spectroscopy (XPS): Samples were prepared on cleaned and O₂ plasma treated Si substrates, using same method as given in section 1.2.2.

1.3 Experimental Techniques

1.3.1. Electrical Characterisation: The current-voltage (I-V) characteristics of the devices (active area $= 0.68 \text{ cm}^2$) were studied in N₂ atmosphere, under AM 1.5G 1 sun solar spectrum generated by an ABET 10500 solar simulator (class AAB) with a 150W Xe arc lamp, calibrated to 100 mW cm⁻² with a reference Si cell (Newport, PVM 165). PV devices were connected directly to a Keithley 2400 source measure unit (SMU) as the external load. The devices were illuminated through an aperture mask of 0.433 cm² open area. The EQE characteristics of the test devices were measured using a Bentham PVE300 system under ambient conditions. Devices were biased at short circuit condition and EQE was measured using a dual quartz/halogen light source with illumination wavelengths in the rage of 300 - 1100 nm.

1.3.2. UV-Vis Spectroscopy: The UV-Vis absorption of the LTM thin films were measured using a Varian Cary 5000 UV-Vis-NIR spectrometer fitted with a transmission accessory.

1.3.3. SEM imaging: Prior to SEM measurements, electrical contacts were made using conductive silver ink between the metal SEM stub and the perovskite thin layer. The samples were investigated with a FEI Quanta 200F Environmental SEM setup under a vacuum of $<10^{-5}$ mbar.

1.3.4. OPTP Measurements: The solution processed samples were photoexcited by an optical light pulse at a wavelength of 950 nm (corresponding to a photon energy of 1.31 eV) and then probed by a

terahertz-frequency pulse after a well-defined delay time. The measured change in the terahertz (THz) transmission pulse is proportional to the photoinduced conductivity ($\Delta\sigma$) in the material. The frequency-dependent THz conductivity spectra $\Delta\sigma(\omega)$ is obtained at 60 ps after photoexcitation by scanning the entire transmitted THz waveform and is converted to photoconductivity by $\Delta\sigma(\omega) = \frac{\varepsilon_0 c(1+n_s)}{d_1} \frac{\Delta T}{T}$. The resulting spectra was fitted with the Drude-Lorentz model described by $\Delta\sigma(\omega) = \frac{Ne^2}{m^*} \frac{i\omega}{\omega^2 - \omega_0^2 + i\omega\gamma}$ from which scattering time (1/ γ) was deduced.

1.3.5. GIWAXS Measurements: GIWAXS data was collected using a Xenocs Xeuss 2.0 system with a liquid Gallium MetalJet (Excillum) source with 9.2 keV X-rays. X-rays were incident on perovskite film surfaces under vacuum for scatter reduction. Scattered X-rays from the film were detected using a Pilatus3R 1M detector with a typical sample to detector distance of ~330 mm. GIWAXS data reduction was performed using the GIXSGUI Matlab toolbox, including corrections for solid-angle, efficiency corrections and pattern reshaping.[1]

1.3.6. XPS Experiments: XPS analyses were performed on a ThermoFisher Scientific Instruments (East Grinstead, UK) K-Alpha+ spectrometer. XPS spectra were acquired using a monochromated Al K α X-ray source (hv = 1486.6 eV). An X-ray spot of ~400 µm radius was employed. Survey spectra were acquired employing a Pass Energy of 200 eV. High resolution, core level spectra for all elements were acquired with a Pass Energy of 50 eV. All spectra were charge referenced against the C1s peak at 285 eV to correct for charging effects during acquisition. Quantitative surface chemical analyses were calculated from the high resolution, core level spectra following the removal of a non-linear (Shirley) background. The manufacturers Avantage software was used which incorporates the appropriate sensitivity factors and corrects for the electron energy analyser transmission function.

(SI-2) Grain Size Analysis



Figure S1 | The grain size analysis as calculated from the SEM images shown in Figure 1(b-d) for different anti-solvent treatments where toluene shows the largest grain sizes and the narrowest distribution signifying most uniform grain growth. The average grain sizes are 332.5 ± 21.1 nm for CB, 342.5 ± 29.4 nm for anisole and 387.5 ± 13.4 nm for toluene, showing largest grains for toluene treatment.

(SI-3) Grazing Incidence Wide Angle X-Ray Scattering (GIWAXS) analysis

In Figure S2a scattering planes with Miller indices (100), (110), (111) and (200) are identified associated with the pseudo-cubic perovskite phase, which is similar in all samples. From the lineplots it is clear there is no scattering from secondary phases (such as PbI_2 , which would be expected at $q = 0.89 \text{ Å}^{-1}$) or significant differences in relative intensities of the scattering peaks regardless of the anti-solvent used. *In situ* scattering measurements show that the perovskite in the toluene sample is mostly formed before annealing, (97.0% for toluene compared with 94.8% for anisole and 90.4% for chlorobenzene). This indicates that crystallisation is marginally improved following toluene quenching but prior to annealing; however, the increase in crystallinity for all samples is minimal during annealing (here under vacuum conditions, which may accelerate solvent removal). This makes clear that the crystallisation process is largely induced by the quenching process and following early liberation of any trapped solvent in the as-deposited film the perovskite phase is rapidly formed.



Figure S2 | (a) Reduced 1D X-ray scattering line plots for chlorobenzene, anisole and toluene treated LTM perovskite thin films generated by radially integrating 2D grazing incidence wide angle X-ray scattering (GIWAXS) patterns of annealed films. (it should be noted that peak broadening at high q is due to instrumental broadening and sample area effects,[2] and apparent splitting of the (200) peak is caused by gaps between detector modules.) (b) *In situ* peak intensity tracking for the (100) scattering peak during annealing.

Further crystallographic analysis is performed by considering the azimuthal scattering intensity of the two prominent scattering features from the (100) and (111) planes in Figure S3. This orientation analysis on the films shows comparable texture in each case with intensity maxima of the (100) peak at $\chi = -45^{\circ}$ (45°) and 90° (out-of-plane) corresponding to two dominant crystal orientations. With respect to out-of-plane scattering intensity, toluene has the greatest scattering followed closely by chlorobenzene, with the anisole-quenched sample showing significantly reduced out-of-plane scattering. The chlorobenzene sample has more crystalline domains preferentially oriented at 45° with the highest intensity scattering here, followed by anisole and then toluene. The anisole sample in general shows more isotropic scattering, with broadly higher intensity away from the maximum in the region $-70^{\circ} < \chi < -25^{\circ}$. For the (111) peak the observed maximum is at $\chi \approx 35^{\circ}$ with the chlorobenzene sample showing a small shift to $\chi \approx 37^{\circ}$ and increased intensity of scattering around the azimuthal maximum for this plane. Considering all GIWAXS data, it is clear that overall the anti-solvents have only a small effect on the crystallinity of the perovskite phase and are not enough to cause significant differences in device performance.



Figure S3 | Azimuthal dependence of the (100) and (111) peaks for each anti-solvent treatments. In (a) the (100) scattering detector data is shown for each anti-solvent (after annealing) as a function of χ , the scattering angle relative to the out-of-plane direction. (b) Integrated azimuthal scans across the whole data set (to minimise noise) with data not shown where impacted by gaps between modules in the detector or not captured due to the missing wedge from q_z axis normal to the sample.

(SI-4) X-ray Photoelectron Spectroscopy (XPS) Analysis



Figure S4 | The X-ray photoelectron spectra (XPS) for Sn species on the LTM perovskite surface after the modified solvent engineering process, showing Pb^{2+} peaks, where anisole treated LTMs show highest Pb^{2+} amount while the other two have comparable atomic percentages.

Table S1/ Binding energy (B.E.) and atomic percentage (A%) comparison of the surface Pb and Sn metal cations as obtained by XPS

		Chlorobenzene		Anisole		Toluene	
		B.E. (eV)	A (%)	B.E. (eV)	A (%)	B.E. (eV)	A (%)
Pb ²⁺	Pb(4f _{7/2})	138.18	3.93	138.28	4.59	138.18	3.82
	Pb(4f _{5/2})	143.08		143.18		142.98	
Sn ²⁺	Sn(3d _{5/2})	486.37	4.98	486.45 4	4.22	486.37	4.97
	Sn(3d _{3/2})	494.80		494.89		494.79	
Sn ⁴⁺	Sn(3d _{5/2})	487.55	1.54	487.61	1.54	487.55	1.34
	Sn(3d _{3/2})	496.25		496.33		496.30	

From the atomic percentages given in Table S1, it is clearly observed here that the surface elemental composition is changed according to the anti-solvent used. Considering the atomic ratios of Pb^{2+} and Sn^{2+} on the perovskite surfaces in CB and toluene treated LTM films, it is noted that the surface Pb^{2+}

percentage is lower than Sn^{2+} , giving generally Sn rich perovskite surfaces, whereas the Pb²⁺ atomic percentage in anisole treated samples is higher than that of Sn^{2+} . We note that although the XPS spectra indicate a lower total surface Sn content for the films formed using anisole in comparison to the other two anti-solvents, most of this existing Sn is in the Sn⁴⁺ form.

(SI-5) Optical Pump-Terahertz Probe (OPTP) Measurements

The photoexcitation pulse induced a change ΔE in the transmission of the terahertz (THz) probe pulse. The measured change in transmission is directly proportional to the photoconductivity $\Delta \sigma$ of the LTM perovskites. Both photoconductivity decays and photoconductivity spectra were measured and analysis was performed using the frameworks reported in Ref [3]. The photoconductivity decay dynamics of the three LTM perovskites were observed to be identical (**Figure S5(a)**). Interestingly, it was noted that the photoconductivity peak of the three LTM perovskites were distinct in magnitude with toluene treatment producing the highest photoconductivity followed by CB and ANI respectively. This trend was noted at all measured pump fluences (Figure S5(b)). The mobilities (Figure S5(c)) were extracted from the photoconductivity decays presented in Figure S4(b) by fitting the linear relationship $\mu = \frac{\Delta \sigma}{ne}$ where *n* is the total photoexcited charge carrier density and *e* is the electronic charge. The photoinduced THz conductivity $\Delta \sigma(\omega)$ spectra were well-fitted with a Drude-Lorentz model consisting of a single resonance that accounts for the effect of carrier localisation in discrete grains (Figure 5(d)).



Figure S5 | (a) Photoconductivity decays of toluene, chlorobenzene and anisole treated samples. Decays are normalised and offset for clarity. (b) Peaks of photoconductivity decay vs. pump fluence, showing the highest photoconductivity toluene treated samples followed by the chlorobenzene and anisole, at all measured fluences. (c) The charge carrier mobilities of Pb-only and Pb-Sn perovskites as reported in literature (also determined using OPTP) in comparison with that of the LTM perovskites reported in this work, for different anti-solvents. (d) Real part of the photoconductivity spectra of toluene (red circles), chlorobenzene (black squares) and anisole (blue diamonds) treated samples at 60 ps after photoexcitation at a fluence of $209 \,\mu$ J/cm² and fitted with a Drude-Lorentz model. The lines are fits.

(SI-6) Device Performance of the Pb-Only Analogy of the Perovskite with Difference Anti-Solvents

Pb-only triple cation perovskite (formula; $Cs_{0.05}(FA_{0.83}MA_{0.17})_{0.95}(PbI_{0.83}Br_{0.17})_3$ prepared as reported elsewhere[4]) PV devices with a device stack of FTO/TiO₂/perovskite/Spiro-OMeTAD/MoO₃/Ag (n-i-

p architecture) were fabricated and characterized under ambient conditions. Perovskite layer was casted under N_2 atmosphere using 3 anti-solvents, toluene, anisole and CB were used as anti solvents similar to LTM perovskite given in main text, and annealed at 100°C for 1 h. Unlike LTM PV devices, the Pbonly device parameters depicted in **Figure S6**, does not show a significant variation depending on the anti-solvent used, with values almost lying within error of each other, although CB seems to be giving higher average numbers in all cases (**Table S2**).



Figure S6 | (a) The J-V curves for the champian Pb-only PV devices using different anti-solvents. Box plots of (b) open circuit voltage (V_{oc}), (c) short circuit current density (J_{sc}), (d) fill factor (FF) and (e) power conversion efficiency (PCE) of the devices treated with different anti-solvents. All device parameters appear close to each other within error, not showing much variation for different anti-solvents. In all cases, the best average performance is shown by CB treated devices.

Anti-solvent	Champion PCE (%)	PCE (%)	Jsc (mA cm ⁻²)	V _{oc} (V)	Fill factor (FF)
СВ	19.08	18.14 ± 0.84	22.79 ± 1.15	1.08 ± 0.02	0.74 ± 0.01
ANI	18.19	16.77 ± 0.78	21.95 ± 0.62	1.06 ± 0.02	0.72 ± 0.02
TOL	18.04	16.76 ± 0.74	21.97 ± 0.85	1.06 ± 0.03	0.72 ± 0.02

Table S2 / Device performances of Pb-only perovskite solar cells with different anti-solvent treatments

(SI-7) Hysteresis of LTM devices

The effect of anti-solvents on the hysteresis between forward and reverse scans of the LTM PSC devices were studied under varying scan rates. The hysteresis index calculated as $\frac{PEC_{reverse} - PCE_{forward}}{PCE_{reverse}}$ is given in **Table S3** for each condition. Scan rates of ~0.50 V s⁻¹ best mimic the conditions of the experimental data given in the main text, and it can be clearly seen that toluene treatment has aided in lower hysteresis.

Scan Rate (V s ⁻¹)	Anti-Solvent				
	TOL	СВ	ANI		
0.05	0.049	0.060	0.004		
0.50	0.150	0.366	0.286		
5.00	0.515	0.654	0.542		

Table S3/ Hysteresis Indexes calculated for LTM PSCs

(SI-8) Band Tail Energy Distribution

A measure of the band-tail defect states is quantified by the Urbach energy (E_U) , where a high (low) crystalline order and low (high) disorder is suggested by lower (higher) E_U value. It is noted that, given the E_U depends on the width of the band tail, lower E_U values suggest a narrower band tail energy state distribution.[5] The E_U , for different solvent treatments were calculated using the relationship given by,[6]

$$EQE = IQE \exp\left(\frac{h\nu - E_0}{E_U}\right)$$
(S1)

Where IQE is internal quantum efficiency, hv is the photon energy and E_o is a constant. This can be rearranged to,

$$\ln(EQE) = \left(\frac{1}{E_U}\right)h\nu + \left[\ln(IQE) - \frac{E_0}{E_U}\right]$$
(S2)

Where the linear region of the plot of ln(EQE) against hv (in the band edge) yields a slope of $1/E_U$ (Figure S7(a))



Figure S7 | (a) The Urbach energy (Eu) plots for perovskite thin layers giving the band tail energies estimated by the linear region within the dotted lines, where toluene treated perovskites give the lowest EU. (b) Schematic representation of the distribution of the band tail states for anisole (blue) and toluene (red) as suggested by the Urbach energy and TFL analysis.

(SI-9) Optical Characterization: Band edge and bandgap calculation



Figure S8 | (a) UV-Vis absorption spectra for LTM perovskite thin films treated with different antisolvents. The absorption edges of LTM films are seen at 950 nm (dashed line). (b) The Tauc plots for the aforementioned perovskite films derived from the UV-Vis spectra. The linear regions within the dotted lines were used to estimate the bandgap energies, resulting in a bandgap of ~1.26 eV for all 3 anti-solvents.

Figure S8 depicts the UV-Vis-NIR absorption spectra for LTM perovskite thin layers treated with different anti-solvents, where the absorption band edge is shownby dotted lines around 950 nm indicating that the LTM perovskite is able to absorb longer wavelengths of the solar irradiance spectrum, extending towards the near infra-red region due to their lower bandgaps (which are typically reported to be ~1.2 - 1.3 eV for Pb-Sn mixed perovskites[7]) than Pb only perovskites.[8] To investigate the effect of anti-solvents on the band gap of LTMs and in forming the bulk perovskite crystal structure, a comparison of the bandgap energies (*E*_G) could be derived for the different anti-solvent treatments. A change in *E*_G with the anti-solvent will be reflected as a shift in the optical absorption edge of the corresponding perovskite film, which results from shifts in conduction and/or valance band positions. In order to calculate the bandgaps for the LTM films, further analysis was carried out using the Tauc relationship[9], [10] given below,

$$(\alpha h v)^{\frac{1}{n}} = \beta (h v - E_{BG}) \tag{S3}$$

Here α is the absorption coefficient of the material, β is the band tailing parameter, and *n* is the power factor of the transition mode which is governed by the nature of the electron transition depending on the band structure of the material and whether photon-assisted transition is required (*n* = 0.5, 1.5, 2, and 3 for direct allowed, direct forbidden, indirect allowed, and indirect forbidden transitions respectively).[11] Given that both Pb and Sn halide perovskites have been reported to be direct bandgap materials,^{40,41} *n* = 0.5 (direct allowed transition) and α of the material can be given by,[15]

$$A = \alpha \times l \tag{S4}$$

where l is the optical path length. Therefore, after rearrangement of terms, Eqn.S4 becomes,

$$(Ah\nu)^2 = \beta l^2 h\nu - \beta l^2 E_{BG}$$
(S5)

where the intercept divided by the slope in the linear region of the plot of Ahv^2 vs. hv directly gives the E_G of the material. These were calculated to be ~1.26 eV irrespective of the anti-solvent used which is in excellent agreement with the LTM perovskites reported in literature[6]. Therefore, it can be concluded that the average composition of the bulk phase is similar in all films.

(SI-10) Dark Current Analysis

Further in-depth analysis of the dark current characteristics indicates the existence of regions with different slopes (*m*), where current density (*J*) changes accordingly with the voltage (*V*) following the power law $J \propto V^m$.[16] By careful inspection of the dark curves, it can be noted that there are regions of different slopes (*m*). Generally in diodes at low electric fields (low voltages), the *J* vs *V* relationship obeys Ohm's law:

$$J_{\Omega} = ne\mu \frac{V}{d}$$
(S6)

Where *n* is the charge carrier density, *e* is electric charge, μ is charge carrier mobility and *d* is the semiconductor thickness. At low (< 1 V μ m⁻¹) electric fields, in which the low current behaviour of diodes is observed, the *J*-*V* characteristics are said to be governed by the interfaces of the diode.[17] When the electric field is increased into the high electric field regime (> 1 V μ m⁻¹), the transport properties of charge carriers are dominated by the properties of the semiconductor. In an ideal trap free case, the *J*-*V* relationship follows the space charge limited current (SCLC) mechanism according to Child's law[17]:

$$J_{SCLC_{trap\,free}} = \frac{9}{8} \varepsilon \varepsilon_0 \mu \frac{V^2}{d^3} \tag{S7}$$

where ε and ε_0 are the relative permittivity of the semiconductor and the permittivity of free space respectively.

For the case where the semiconductor is incorporated with shallow traps, Child's law is modified to [17]:

$$J_{SCLC \ shallow \ traps} = \frac{9}{8} \varepsilon \varepsilon_0 \theta \mu \frac{V^2}{d^3}$$
(S8)

Where,

$$\theta = \frac{n_m}{n_m + n_t} \tag{S9}$$

Where n_m and n_t are mobile and trapped charge carrier densities respectively. Here, both these charge types contribute to J_{SCLC} and the slope of the *J*-*V* curve is dependent on these charge carrier densities. Furthermore, shallow traps which are located between the Fermi energy and the band edge, are known to decrease the photocurrent density of a solar cell.[18] When shallow traps are present, the Ohmic regime is prolonged due to charge trapping (resulting in very low n_m and giving rise to quasi-Ohmic behaviour), and the gradient increases gradually when $\theta \rightarrow 1$ with increasing electric field in the SCLC regime (in which, $J \propto V^2$).[17] With increasing electric field, the diode in operation reaches the trap filled limit (TFL) at $V=V_{TFL}$, when the majority of trap sites are filled, hence the current can flow freely, thereby noticeably increasing the total current.[19] The current density in the trap filled limit is given

by Eqn.S10, where *B* and *l* are constants with the constant *l* relating to the steepness of the distribution of trap states[19] as depicted in Figure S7(b) (where $l_1 > l_2$).

$$J_{TFL} = B \frac{V^{l+1}}{d^{2l+1}}$$
(S10)

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