# Back-Contact Perovskite Solar Cells

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Interdigitated back-contact (IBC) architectures are the best performing technology in crystalline Si (c-Si) photovoltaics (PV). Although single junction perovskite solar cells have now surpassed 23% efficiency, most of the research has mainly focussed on planar and mesostructured architectures. The number of studies involving IBC devices is still limited and the proposed architectures are unfeasible for large scale manufacturing. Here we discuss the importance of IBC solar cells as a powerful tool for investigating the fundamental working mechanisms of perovskite materials. We show a detailed fabrication protocol for IBC perovskite devices that does not involve photolithography and metal evaporation.

# Introduction

Metal halide perovskite solar cells represent one of the most promising photovoltaic technologies to date, <sup>1,2</sup> with power conversion efficiencies (PCEs) exceeding 23% for single junction cells<sup>3,4</sup> and 28% for perovskite on silicon tandem solar cells<sup>5,6</sup>. Other applications include light-emitting diodes, <sup>7-10</sup> lasers, <sup>11,12</sup> and photodetectors<sup>13,14</sup>. This work focusses on interdigitated back-contact (IBC) perovskite solar cells, which have enabled the discovery of photon recycling in perovskite materials and facilitated simultaneous *in-situ* and *in-operando* studies of perovskite film formation. <sup>15,16</sup> In our recent work, which is discussed here by Mejd Alsari and Mojtaba Abdi-Jalebi, we reported relatively high open circuit voltage in IBC perovskite solar cells during the early stages of the anneal. This suggests that not fully formed perovskite films display clean semiconducting properties and defect tolerance, which is discussed here by Richard Friend and Luis Pazos. We conclude the introduction with a conversation on photon recycling by Felix Deschler and Luis Pazos. Next, Mejd Alsari illustrates a detailed fabrication protocol of IBC perovskite solar cells. Finally, Felix Deschler and Luis Pazos wrap up this work with a discussion on future developments of IBC perovskite solar cells.

# **Defect Tolerance**

The most widespread, flexible, and low-cost deposition technique for perovskite materials is solution processing.<sup>5</sup> In the simplest ABX<sub>3</sub> perovskite formulation, an organic halide salt AX (e.g.  $CH_3NH_3X$  or  $CH(NH_2)_2X$ , X = I, Br, Cl) and a heavy metal salt  $BX_2$  (e.g.  $PbX_2$ , X = I, Br, Cl) are mixed in a polar solvent (e.g. dimethylformamide, dimethyl sulfoxide) to form a perovskite precursor solution.<sup>1,17-24</sup> Higher device efficiencies and longer stability can be obtained by mixing multiple ions at the A and X sites.<sup>1,25-27</sup> The perovskite precursor ink can be deposited at room temperature with a variety of techniques, including spin-coating,<sup>28</sup> spray-coating,<sup>29</sup> inkjet-printing,<sup>30</sup> and slot-dye coating<sup>31</sup>. Deposition is typically followed by post-processing treatments, such as thermal annealing. Solution processing is intrinsically 'fast and messy' and does not lead to the formation of single crystals but rather polycrystalline films.<sup>32-35</sup>

Due to the incredible amount of impurities and structural defects caused by solution processing, polycrystalline perovskite films have a very high trap density ( $^{\sim}10^{16}$  cm $^{-3}$ ).  $^{34,36,37}$  If you look at consolidated device physics and electronics theories, which are taught at undergraduate university courses, you would think that devices based on solution processed perovskite could not possibly work. In fact solar cells based on GaAs with analogous levels of doping would not work at all.  $^{38}$  However, perovskites surprisingly behave as well as standard inorganic intrinsic semiconductors and solar cells based on these materials have been described as p-i-n (or n-i-p) devices.  $^{38,39}$ 

Perovskites such as methylammonium lead iodide (MAPbI<sub>3</sub>, MA = CH<sub>3</sub>NH<sub>3</sub><sup>+</sup>) display exceptional optoelectronic properties comparable to GaAs. <sup>2,20,40,41</sup> These include strong light absorption, <sup>42-44</sup> non-exicitonic behaviour, <sup>42,45,46</sup> low recombination rates, <sup>47</sup> long carrier lifetimes (> 1  $\mu$ s) <sup>36</sup> and diffusion lengths (>1  $\mu$ m) <sup>40,48,49</sup>, and photon recycling <sup>16</sup>. Further similarities with inorganic semiconductors include relatively small carrier effective mass, <sup>50,51</sup> very sharp diffraction rings (*cf.* polycrystalline materials), <sup>15,35,52</sup> and conduction (or valence) band dominated by inorganic cations (or anions) <sup>50,51,53</sup>.

In spite of these similarities, charge-carrier mobilities (<100 cm²V⁻¹s⁻¹) are significantly lower than those of GaAs (~8000 and ~400 cm²V⁻¹s⁻¹ for electrons and holes, respectively). <sup>2,41</sup> As suggested by the mobility trend vs temperature ( $\mu \propto T^{-3/2}$ ), this could be due to electron-lattice coupling effects (acoustic phonon scattering), rather than impurity scattering. <sup>41,54-58</sup> Nevertheless, devices show high open circuit voltage ( $V_{oc} > 1.0 \text{ V}$ ) and with a bandgap ( $E_g$ ) of ~1.5eV the  $V_{oc}/E_g$  ratio is very similar to that of GaAs thin film solar cells. <sup>59</sup> These aspects contribute to very high PCE in perovskite solar cells, indicating that these materials are inherently defect tolerant in spite of point defects, dislocations, stacking faults, grain boundaries, or surfaces and behave as a clean semiconductor (without defects or impurities). <sup>60-62</sup>

Although the exact origin and nature of traps in perovskites is still under debate, it is believed that intrinsic defects mainly result in shallow traps (close to the band edges), which might provide an explanation for the long carriers diffusion length and high V<sub>oc</sub> in solar cells.<sup>59,63</sup> Charge carriers are probably unaffected by defects thanks to polaron screening.<sup>55,64</sup> Defect tolerance might be the result of a bonding conduction band minimum (CBM) and an antibonding valence band maximum (VBM). Here dangling bonds caused by a vacancy can resonate inside the bands, leaving the band gap free of deep states.<sup>62,65,66</sup> Moreover continuous exposure to light can cause photo-induced ion migration and a reduction in trap density in the perovskite film (photo-brightening).<sup>67</sup>

On the contrary low  $V_{oc}$  can be a symptom of deep traps (within the bandgap), which cause Shockley-Read-Hall non-radiative recombination centers. <sup>59,68</sup> In MAPbI<sub>3</sub> deep traps could arise from the formation of Pb dimers and I trimers, <sup>69</sup> and/or I dimers <sup>70</sup>. A recent work by Meggiolano *et al.* suggests that only less abundant interstitial iodine defects introduce deep carrier traps in MAPbI<sub>3</sub>. <sup>61</sup> Electron traps can be neutralized thanks to the iodine redox chemistry leaving only short-living hole traps as 'killer' defects, <sup>61</sup> which can be converted into inactive electron traps under mild oxidizing conditions or by doping the perovskite (e.g. with Cl or Br). <sup>61</sup> Interstitial iodine can also be minimized with a slightly under stoichiometric I: Pb (~2.8:1) ratio. <sup>61</sup>

Further reducing deep-traps would further maximize the  $V_{oc}$  and ensure non-radiative decay is eliminated.<sup>71,72</sup> At this point all charges would only recombine through radiation of light (i.e. a great solar cell is also a great light-emitting diode).<sup>1,72</sup> As will be discussed later, this light can be then

'recycled' by the solar cell, thus contributing to improved charge transport and  $V_{oc}$ , or lost outside the cell by luminescence.<sup>1,16</sup>

In the following section, Luis Pazos and Richard Friend talk about defect tolerance in perovskite materials.

**Luis M. Pazos-Outón (LMPO):** One of the key challenges for the new semiconductor technologies is to achieve a good passivation strategy and, therefore, get rid of traps.<sup>73-75</sup> Do you think there is anything in perovskites that allow them to have such high performance?

**Richard H. Friend (RHF):** This is probably the core of why the perovskites turn out to be so surprising. In spite of the fact that they have many structural and chemical defects, due to the way they are made and the level of impurities that are present, the luminescence, which is the most sensitive test, remains extremely efficient and that is unprecedented. Other non-crystalline thin film semiconductors generate really low luminescence yields. We start with a surprise, no matter how you fabricate the lead halide perovskite film, the perovskite is actually very luminescent. All sorts of approaches are being taken in the literature to improve on it by passivation, that is removing defects states that quench luminescence around the crystallites, maybe in the bulk, but almost the surfaces are being cleaned up with these approaches. 16,78-83

**LMPO:** To achieve a very high luminescence you certainly benefit from having very low trap densities and low Shockley-Read-Hall (SRH) recombination rates, but also having a very strong radiative rate is beneficial.<sup>84</sup> Do you have any explanation of why the radiative rate is so strong?

**RHF**: The very strong radiative recombination is again something that we find surprising. The recombination, where the electron and hole come together again, we know that the kinetics are bimolecular, so the rate depends on the electron concentration times the hole concentration. Usually that does not give a very efficient luminescence, because those collisions do not happen all that quickly, but when they do, they certainly produce photons. It hink the key is to ratio the radiative recombination processes against everything else going on, which can quench luminescence. It is again surprising that the non-radiative channels are relatively unimportant.

**LMPO:** A work published in Nature Photonics in 2016 on laser cooling<sup>87</sup> showed an external luminescence efficiency of 99.8% and that is really extraordinary. That was on a single crystal perovskite. If they have achieved that in a single crystal, do you think that perhaps the field should move from solution processed materials that are polycrystalline and have higher defects, to single crystals?

**RHF:** The reality is that we are not going to be able to this. If we want to make solar cells because we need to have film absorbing materials that are quite thin of the order of a fraction of a micron. Unless someone finds a remarkably easy way of growing thin crystals *in-situ*, that is not going to happen. Back to the surprise, which is that the performance of polycrystalline materials turns out to be remarkably good and that there appear to be methods to improve on the original recipes, of course the other factor that comes into play when we make a device, which might be a light emitting diode or a solar cell, is that the luminescence can easily be quenched at the interface the semiconductor and the perovskite makes with the two electrodes for electron and hole management and the current perovskite literature shows that there is still some quenching. There

are some chemical defects, whatever chemistry there is at the interface (i.e. with  $TiO_2$  for electron extraction) it is possible to apparently produce energy states that lie in the bandgap and there is some quenching. So in respect of whether it is a single crystal or a thin film which is polycrystalline, I think a lot of the future efforts are going to be to understand that interfacial chemistry to be able to remove quenching there. We tend at the moment to concentrate more on the pure material. As the field becomes more developed, we will pay more attention at what happens at the interfaces.

**LMPO:** What do you think that is actually helping to passivate the defects?

**RHF:** The passivation appears to be something that can be done with almost any of the compositions. If one is looking at the sort of fundamental issues, the electronic structure, the band structure is really determined by the Pb and the Br, and I and the third cation probably does not do a lot more than control the lattice parameter. So I wouldn't expect to see a fundamental difference as we tune the composition of the cation. I know it is advantageous for other reasons. But your work rather elegantly brings into play that we have to look at that as we had two operating devices at higher brightness and under concentration the non-radiative decay of electrons and holes through Auger processes becomes quite important. Something which I think I have learned from you that I had not appreciated which is the even Si has its performance limited by Auger processes.

**LMPO:** Generally in most semiconductors one of the key requirements is to be able to dope the material effectively. However for perovskites this seems problematic.<sup>60</sup> I was wondering if anyone in your group has tried anything along this direction.

**RHF:** The interesting thing about perovskites is that they break just about every rule and they work very well. They are much cleaner than they should be. They are very hard to dope and most semiconductor technologies have depended on the ability to extrinsic p-type or n-type dope. That is often critically important for getting a really good quality Ohmic electrode. For organic semiconductors, doping has been engineered after a few decades of hard work and is alive and well in the OLED displays.<sup>89</sup> For the perovskites it may well be that the ability of the perovskite materials to adjust itself locally, to neutralize the effect of a defect, which we suspect is why they are so luminescent and so clean, will also mean that you cannot dope them. If you try to put a wrong valence cation in, the local chemistry will adjust itself and you will not create the doping. The literature really suggests that may well be the case. We have done a lot of work putting in monovalent cations that in principle might substitute for the Pb. 90 Those materials rather than being extrinsically doped are even less doped than the materials by themselves. Moreover, the literature on perovskite field effect transistor is relatively small.<sup>91-94</sup> When it is possible to see a field effect, what we are not sure about is whether that field effect eventually is reduced because the induced charges can be compensated by structural rearrangement around the charge that has the effect of screening it away.

**LMPO:** It is really interesting that the same process that prevents doping also allows you to not to need doping.

**RHF:** I think that is the case. I have not seen a lot of explicit discussion of that in the literature but I think that is the case.

# Interdigitated Back-Contact Solar Cells

In the following section, Mejd Alsari and Mojtaba Abdi-Jalebi talk about how interdigitated back-contact (IBC) devices have enabled exploring the correlation between the structural and opto-electrical properties of perovskite solar cells.

Mojtaba Abdi-Jalebi (MA-J): The commonly deployed solar cell architecture is represented by a mesoscopic or planar heterojunction (see Figure 1a). 95,96 The perovskite film, which is several hundred nanometer thick, serves as the intrinsic semiconductor absorber layer, and is sandwiched between an electron-transporting material (ETM, n-type or hole-blocking layer), and a hole-transporting material (HTM, p-type or electron-blocking layer). 97 These carrier-transporting materials serve as electrode buffers for selective carrier extraction. In this device configuration, typically a transparent conducting oxide (TCO) layer such as indium tin oxide (ITO) or fluorine-doped tin oxide (FTO) must be used for at least one of the electrodes. Moreover, multiple coating steps are required to produce a device with good charge carrier extraction efficiency. This includes the deposition of the absorber layer, buffer interlayers, and contacts. In perovskite solar cells, the illumination takes place from the TCO side (bottom illumination). On the other hand the main architecture for the market leading technology crystalline Si is interdigitated back-contact solar cell (IBC, see Figure 1b). 98,99 Mejd, can you explain a bit about this IBC architecture?

**Mejd Alsari (MA):** In the IBC architecture the electron- and hole-selective electrodes are copositioned on the backside of the cell in an interdigitated fashion. The IBC solar cell is completed by depositing the active layer on the interdigitated electrodes. With this architecture, optical transmission losses caused by the top contact can be avoided, through the possibility to illuminate the solar cell from the absorber layer side (top illumination). Because electrodes transparency is not required, a wide variety of electrode materials can be employed, without compromising conductivity (resistive losses). In addition, such architectures offer the possibility to maintain the interconnection circuitry on the rear side of the cells on one common surface, facilitating solar cell module assembly processes. This concept was originally developed for concentrating photovoltaics by Schwartz and Lammert in 1975, and has been successfully utilized in Si-based, 101-104 CdTe, 105,106 dye-sensitized, 107,108 organic, 109 and most recently perovskite solar cells.

MA-J: What has been done so far with IBC perovskite solar cells?

MA: There have been only a few reports on IBC solar cells employing perovskites, some focussed on device optimization<sup>111-114</sup> and others focussed on understanding the device physics of perovskite materials<sup>15,16</sup>. In addition to the advantages mentioned above, IBC devices have been proposed by Bach's group as a potential solution to overcome issues related to pin holes (i.e. shorting between electrodes) and mitigating damages of the perovskite layer during the top electrode deposition.<sup>111</sup> In 2016 this group proposed the concept of the quasi-interdigitated electrodes architecture (see Figure 1c) and reported stable MAPbl<sub>3</sub> devices with a PCE of 3.2%.<sup>112</sup> The drawback of this architecture is that it involves rather complicated photolithography and deposition steps, which is a bottleneck for large scale production.<sup>113</sup>

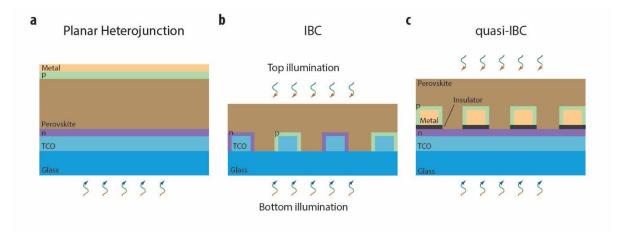


Figure 1 | Illustration of three perovskite solar cell architectures. The planar heterojunction in **a** is the 'direct' architecture. The n and p-layers can be swapped to obtain an 'inverted' architecture. **b**, Interdigitated Back-Contact architecture. **c**, quasi-Interdigitated Back Contact architecture.

Hu *et al.* reported a different back-contact perovskite solar cell architecture achieving 3.9% PCE. <sup>113</sup> The authors do not employ any TCO nor any hole selective layer. However, this architecture involves several fabrication steps, including spin-coating and annealing TiO<sub>2</sub>, sputtering Ti, spin-coating and thermally oxidizing ZrO<sub>2</sub>, a two step MAPbI<sub>3</sub> perovskite deposition, and a top Au electrode evaporation.

Recently, Jumabekov *et al.* developed a back-contact architecture (quasi-integrated electrodes) based on modified 'natural lithography'.<sup>114</sup> If we exclude TCO and top electrode, this architecture does not involve any other traditional lithographic step. The TCO substrate is functionalized with self-assembly monolayers, which act as a sacrificial lithographic mask for a monolayer of polystyrene microbeads. A cathode made of Al<sub>2</sub>O<sub>3</sub>/Al/NiCo is then evaporated. The mask is subsequently etched. After an annealing step to form oxide shells around the exposed metalling components of the cathode, MAPbl<sub>3</sub> is spincoated to complete the device. Although this work simplifies the earlier architecture reported by the same group<sup>112</sup>, the resulting PCE is relatively low (1.75%). The same group proposed another variant to this architecture and achieved a maximum PCE of 5.39% with MAPbl<sub>3</sub>. This device structure is based on honeycomb-like charge collecting electrodes, but it introduces further complications involving photolithography and e-beam evaporation, which are probably unsuitable for large scale production.<sup>111</sup>

MA-J: Can you talk about how the IBC architecture was used in our recent publication?<sup>15</sup>

**MA:** In our two recent studies we use the IBC perovskite structure to investigate the charge dynamics and as well as to investigate the correlations between the structural and opto-electrical properties of these solar cells during film formation and in these two studies we use a simple IBC structure. <sup>15,16</sup> For the second study we used a structure, where we have commercially available interdigitated ITO substrates. <sup>15</sup> We also use electrodeposited titanium dioxide, electro-polymerised PEDOT, and spin coated perovskite.

One of the advantages of using this architecture is that the perovskite deposition represents the last fabrication step. Because the absorber layer is not blocked by any other layer, it can be directly probed during solar cell operation, and that can be coupled with many investigation techniques such as X-ray diffraction, photocurrent and photo luminescence mapping, etc.

MA-J: Can you talk in more details about the work that had been done at the ESRF?

**MA:** In ref.<sup>15</sup> we use the IBC architecture to investigate the structural and photovoltaic properties of perovskite solar cells during annealing, which is the last stage of the device fabrication. Such study would not have been possible without the IBC structure. As the precursor solution is spin-coated on the IBC substrate, the so-called precursor film displays a clean semiconducting performance since the early stages of the anneal, as highlighted by simultaneous *in-situ* synchrotron X-ray and *in-operando* current-voltage measurements.<sup>15</sup> We observe a peak in the  $V_{oc}$  before the precursor fully converts into perovskite, probably due to a favourable combination between the band gaps of precursor and perovskite phase.<sup>15</sup> We find analogous trends between the perovskite peak intensity extracted from GI-WAXS and the PCE. These trends are confirmed by similar profiles in the figures-of-merit of planar heterojunction solar cells annealed for different time periods and tested *ex-situ*. Overall this work reveals important relationships between the structural properties and device performance of solar cells during film formation while suggesting that defect tolerance is already apparent in the precursor film and evolves during its conversion into fully crystallised perovskite.

## Photon Recycling

In this section, Felix Deschler and Luis Pazos discuss photon recycling in perovskite materials. In ref.  $^{16}$  we performed simultaneous confocal photoluminescence and photocurrent mapping on IBC perovskite solar cells with channel lengths of 4  $\mu$ m. These studies clarify the nature of the long carrier diffusion length, which is enhanced by photon recycling. Photon recycling is a well-known phenomenon in GaAs solar cells, and refers to the fact that when charges recombine radiatively the emitted photons can be reabsorbed (i.e. recycled) by the perovskite.  $^{16}$ 

**Luis M. Pazos-Outón (LMPO):** Felix, can you explain some of the basic concepts of photon recycling in perovskites?

**Felix Deschler (FD):** When we studied the photoluminescence in these metal halide perovskites, initially we found that they are very bright materials. The luminescence also overlaps quite a lot with the absorption edge of the materials. With this combination of factors you could see this photon recycling effect where you generate charge pairs by absorbing a photon in the material and then the recombination of this charge pair generates another photon in the material that does not out-couple directly, does not leave the material directly, but gets reabsorbed and then generates another electron-hole pair, and by this sequence of processes you get a photon recycling effect. When we tried to see this together in our paper, we used photoluminescence microscopy to look at the transport of charges and photons in the material and found that the transport occurs at length scales that are beyond the expected length scales from just absorption and we were really excited to see that. We then concluded that photon recycling has to happen in this case. It was very exciting to see this effect because it is relevant for devices.

**LMPO:** You would need to have a very accurate measurement of the recombination rates. You need to know how much is going into recombination and how much is going into SRH. You have done quite a lot of work on that. How have you done those measurements?

**FD:** In order to really record the recombination process of excited charge carriers of electrons and holes in the perovskite we use ultrafast laser spectroscopy techniques to study what happens when we use fs pulses to put a certain number of carriers into the material and how do they relax into

equilibrium conditions in the dark. These fs pulses that we use are generated in large laser systems, so-called amplifiers, for which this year (2018) the Nobel Prize was awarded (to Gérard Mourou and Donna Strickland for their invention of chirped pulse amplification). We use a technique called pump and probe spectroscopy, where we measure the absorption of light through the material. If we have excited states in the material we get a little bit more light through the perovskites.

That was especially relevant for this photon recycling and photoluminescence yield because when we did our measurements, they did not agree with the quantum yield we measured externally and so we realized some other process has to happen and that allows us to quantify the amount of photon recycling in the material.

#### Protocol

In this section we describe the protocol for the fabrication of IBC perovskite solar cells, following the steps of Alsari  $et~al.^{15}$  In Pazos et~al. we patterned the ITO substrates in our laboratory due to the unavailability of commercial  $4\mu m$  gap interdigitated ITO substrates. This was a time-consuming procedure involving photolithographic patterning. The patterning needed to be very precise to avoid shunting between the hole- and electron-selective electrodes. We thought that such a small gap was required based on the assumption that for efficient charge carrier extraction the interdigitated electrodes must be spaced no more than the charge carrier diffusion length of the active material, which for perovskites is in the order of a few microns. A8,116,117 However, in Alsari  $et~al.^{15}$  we demonstrate that larger gaps can be used and, in fact, commercially available pre-patterned ITO substrates with electrode spacing ranging from 50  $\mu$ m up to 200  $\mu$ m worked as well as the substrates with 4  $\mu$ m gap, when employed in the fabrication of solar cells.

Electron- and hole-selective layers of  $TiO_2$  and poly-3,4-ethylenedioxythiophene (PEDOT) were respectively electrodeposited and electro-polymerised on the interdigitated ITO electrodes. Although various methods can be used to deposit such materials, we preferred electrodeposition (ED) over other techniques due to its low process temperature, ability to control composition and morphology by electrochemical parameters, convenience of depositing films on complex structures, and low cost of raw materials and equipment.  $^{118}$ 

## **ITO Substrates**

- 1. We purchased interdigitated ITO substrates from Ossila Ltd (code S162). We chose the '>50 micron variable length (100 pack)' substrates, which in November 2018 are listed for £425 (i.e. £4.25). This is negligible compared to the (labour) costs involved in the photolithography step that we followed for the work reported in Pazos et al. These prepatterned ITO OFET substrates were actually designed to enable the fabrication and characterisation of transistors without the need for vacuum evaporations or probe stations.
- 2. The substrates need to be cleaned before use.
- 3. Place the substrates on a substrate holder and immerse them in a beaker filled with acetone.
- 4. Place the beaker in a sonicator filled with water and sonicate for 10min.
- 5. Transfer the substrate holder to another beaker containing isopropanol and sonicate again for 10min.
- 6. Dry the substrates with nitrogen stream.

# TiO<sub>2</sub> Electrodeposition

We cathodically electrodeposit TiO<sub>2</sub> from an aqueous peroxo-titanium solution. <sup>15,16,119</sup>

#### **Electrodes**

- 1. Place a 25mL glass beaker under a retort stand.
- 2. Connect a 5cm silver wire to a small crocodile clip, connect the clip to a long aluminium wire, and mount this on the stand. This is the reference electrode.
- 3. Repeat the step above for a platinum foil, which is the counter electrode.
- 4. Connect one side of the substrate to electrical connection legs for ITO glass substrates ('electrical connection legs for ITO Glass Substrates (for 1.1mm glass)', E241, Ossila).
- Then bend the leg so that it fits in the beaker and wrap it with Parafilm (to avoid the connection legs touching the solution). Mount the ITO substrate on the retort stand. This is the working electrode.
- 6. Connect the three electrodes to the Autolab galvanostat.

#### Galvanostat

- 1. Turn on the galvanostat and open the GPES software.
- 2. Go to Method -> Chrono methods -> Amperometry.
- 3. In the Manual Control popup select a current range of 1mA.
- 4. Type a potential of -1.05V, a duration of 800s, and a sampling time of 0.05s.

#### Solution

- 1. Pour 20mL of deionized water into a 50mL centrifuge tube.
- 2. Weigh 64mg of titanium (IV) oxysulfate salt (TiOSO<sub>4</sub>) (14023-100G, Sigma-Aldrich) with a precision scales and transfer it to the centrifuge tube.
- 3. Add 366 $\mu$ L of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) (H/1750/15, Fisher Scientific) to the centrifuge tube.
- 4. Mix the solution with a centrifuge.
- 5. Weigh 202.2mg of potassium nitrate salt (KNO<sub>3</sub>) (221295-100G, Sigma-Aldrich) and transfer it to the centrifuge tube.
- 6. Pour the solution into the beaker.

# Deposition

- 1. In the galvanostat control software press START.
- 2. During the deposition monitor the current vs time curve, which give an indication of how good the deposition is. If the curve is flat there is not any deposition taking place.
- 3. Rinse the substrate with water and ethanol, then dry it with N<sub>2</sub> stream.
- 4. Inspect the substrate and make sure the deposited titanium dioxide is uniform across the substrate (for homogenous films, the solution must be cooled and kept at 10°C during deposition).
- 5. Anneal the substrate using a hotplate at 300°C for 1h in ambient conditions.

#### PEDOT Electropolymerisation

We deposit PEDOT through electropolymerisation. 15,16,120

## Electrodes

1. After cleaning the silver and platinum electrodes, place them back to the previous configuration in a clean beaker.

2. Connect the other side of the ITO substrate to a new electrical connection leg.

#### Galvanostat

1. Go to Methods -> Chrono Methods -> Amperometry and type a potential of 1.25V, a duration of 1s, and a sampling time of 0.01s.

#### Solution

- 1. Pour 20mL of propylene carbonate (CH₃C₂H₃O₂CO, 82229, Sigma-Aldrich) into a 50mL glass vial.
- 2. Weigh 2.12g of lithium perchlorate (LiClO<sub>4</sub>, 634565, Sigma-Aldrich) and transfer it into the vial.
- 3. Sonicate the vial with the sonicator for 30min.
- 4. EDOT is toxic and should be handled in a fume hood. Move the vial to a fume hood and add 21.4µL of EDOT (483028-10G, Sigma-Aldrich).
- 5. Close the vial and shake it.
- 6. Pour the solution into the beaker.

### Deposition

- 1. In the galvanostat control software press START.
- 2. During the deposition monitor the current vs time curve as above.
- 3. Remove the substrate from the electrical connection legs.
- 4. After completion, rinse the substrate with acetonitrile and dry it with N₂ stream.
- 5. Visually inspect the substrate and make sure PEDOT is uniform across the substrate.

## **Active Layer Deposition**

The triple cation perovskite  $Cs_{0.5}(MA_{0.17}FA_{0.83})_{0.95}Pb(I_{0.83}Br_{0.17})_3$  precursor solution was prepared by dissolving 1M FAI, 1.1 M PbI<sub>2</sub>, 0.2M MABr, 0.2M PbBr<sub>2</sub> in a 4:1 (v:v) mixture (organic cations were purchased from Dyesol and the lead compounds from TCI) of anhydrous DMF:DMSO and 1.5M stock solution of CsI in DMSO was added (DMF and CsI were purchased from Sigma Aldrich) to above solution in 5:95 volume ratio. <sup>121</sup> The perovskite solution was then spin-coated at 6000 rpm 30s under ambient conditions. During the second step,  $50\mu$ L of chlorobenzene was dripped on the spinning substrate 10s prior to the end of the program.

#### Solar Cell Measurements

- 1. Turn on the solar simulator and let it warm up. Set the irradiance to 1 sun.
- 2. Connect the IBC solar cell electrodes corresponding to the  $100\mu m$  gap to the multimeter using electrical connection legs.
- 3. Set the following sweep parameters: sweep range from -0.2V to 1V, voltage step 10mV, and 50ms delay.

## Conclusion

In the last part of this work, Felix Deschler and Luis Pazos discuss the future developments of IBC perovskite solar cells at the University of Cambridge.

**Luis M. Pazos-Outón (LMPO):** Can you discuss about your current project on back-contact perovskite solar cells.

**Felix Deschler (FD):** Yes, it is a very interesting structure where we basically take our vertical device and we flip it over of 90° and try to make all the electrodes *in-plane*. That helps us to make the solar cell more transparent because in this way we can minimize the area of the reflective electrodes. Potentially they could be quite good for applications such as windows, so wherever you want to transmit some light through the solar cell. Additionally it gives us access to the surface of the perovskite film, which is usually the most defective area. Now we have a surface that we can address, we can use chemical passivation and other fabrication techniques to reduce the number of defects that we have on this dominant surface area of the perovskite film.

LMPO: You can use passivation agents that are not even conductive, right?

**FD:** Exactly, we do not need to care about extracting charges from this interfaces, which is usually in the vertical device is the largest interface that you need to get charges through. We tried already a few approaches and it seems that we can efficiently passivate these films very well and get high luminescence yields in them. Now we are trying to combine this all to make one big structure and then try with photonic structures on top to modulate the photon density internally so that we can boost the photon recycling effect. Particularly something that is very nice about the back contact is that the surface structure does not have to be functional. We do not have to put a structured electrode on top, you can just put some non-conductive dielectric stack on top that has a photonic bandgap in the right energy range and then hopefully we will improve the efficiency of the devices.

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#### Conflicts of Interest

The authors have no conflicts of interest to declare.

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# **Author Contributions**

M.A. and M.A.J. fabricated and measured the IBC devices. M.A. wrote the manuscript. All authors contributed to this work.

# Keywords

Perovskite solar cells, interdigitated back-contact solar cells, perovskite photovoltaics, perovskite pv, clean semiconductor, defect tolerance.

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