Supporting Information

Monolithic Perovskite/Silicon-Heterojunction Tandem Solar Cells

Processed at Low Temperature

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This supporting information contains the following:

- JV characteristics of perovskite single junctions after ITO deposition
- JV characteristics of perovskite and tandem cells after 3 months storage
- EQE spectra and reflection with and without AR coating
- Current and power output transients
- Simulated reflectance and absorbance as function of ITO thickness
- Simulated reflectance and absorbance as function of the refractive index of ITO
- Simulated reflectance and absorbance for ITO or µc-Si as recombination contact

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FIG. S1: (a) JV characteristics of a perovskite single junction reference cells on glass/ITO substrates with opaque gold electrodes. On top of the gold electrode, MoO₃ and ITO were deposited in the same way as for tandem devices. The JV characteristics shows, that ca. 30 mV loss in V_{oc} can be measured after the MoO₃ and ITO sputter deposition. This is either caused by the thermal stress when evaporating MoO₃ or caused by high energy ions/electrons and or UV irradiation during the sputter process.



FIG. S2: (a) *JV* characteristics of a perovskite single junction reference cell with glass/ITO substrates and opaque gold electrodes measured with a scan rate of 500mV/sec in the first week after sample preparation and after the samples were stored unencapsulated for 3 or 4.5 months in a nitrogen filled glove-box. Note that no temperature control of the device during measurement could be applied. Also, no aperture was used during this measurement and thus, edge effects can cause overestimation of the measured current density. Overall the performance is very stable with slightly decreased J_{sc} and FF but enhanced V_{oc}. This results in a relative efficiency loss of ca. 4% over a period of 3 months and 6% over the period of 4.5 months. The stabilized power output was calculated from the current density measured at a fixed bias of 0.82V over a period of 50 sec and is 11.9%. Thus the stabilized power output is in between the forward and reverse scan with 11.5 and 15.1%, respectively.



FIG. S3: (a) JV characteristics measured under AM 1.5G illumination calibrated to 100 mW/cm² in reverse scan direction (V_{oc} to J_{sc}) of a tandem cell (not a champion device) after preparation and after the sample was stored unencapsulated for three months in a nitrogen filled glove. In accordance with the perovskite single junction reference cells, the performance is very stable with slightly decreased J_{sc} and FF but enhanced Voc. This results in a stable efficiency over a period of three months storage.



FIG. S4: EQE spectra of the champion device with and without AR coating together with the reflection. Indicated is the photocurrent as determined by the multiplication of the EQE spectra with the AM 1.5G spectra and integration over the measured wavelength regime. The AR coating reduces the reflection at 700, 850 and 100 nm. Overall the photocurrent in the perovskite sub-cell is only slightly affected while the photocurrent is enhanced by 1.5 mA/cm² in the silicon sub-cell.



FIG. S5: upper panel: current transients of the champion device measured at different fixed applied voltages ranging from 1.0 to 1.5 V. The dashed line indicates average value of the plateau of the transient which was used for estimating the stabilized efficiency in the main paper. Note that the current density presented in this figure is underestimated caused by shadowing from the contact fingers as explained in the main paper. Lower panel: the efficiency as function of time. Here the current is corrected for the losses caused by the contact fingers. The dashed line at 18.07% indicates the average efficiency deduced from the average current of the plateau as shown in the upper panel.



FIG. S6: (a) Simulated absorbance of the individual sub-cells and 1-reflectance of the tandem device as a function of the ITO thickness in the recombination contact between both sub-cells. With decreasing layer thickness, the reflectance in the wavelength range between 800 and 1000 nm is reduced. Thus the absorbance and with that the EQE of the silicon sub-cell is enhanced. The reflection below 800 nm and with that the absorption of the perovskite is not affected by the ITO thickness in the recombination contact. Details of the simulation can be found in the experimental section.



FIG. S7: (a) Simulated absorbance of the silicon sub-cells and reflectance of the tandem device as a function of the hypothetical ITO refractive index in the recombination contact. The absorption coefficient of the hypothetical ITO layer was not changed. With increasing refractive index, the reflectance in the wavelength range between 800 and 1000 nm is significantly reduced. Thus the absorbance and with that the EQE of the silicon sub-cell is enhanced for standard ITO and a hypothetical ITO having 0.8 higher refractive index, respectively. The reflection below 800 nm and with that the absorption of the perovskite is not affected by the ITO refractive index in the recombination contact. Details of the simulation can be found in the experimental section.



FIG. S8: (a) Simulated absorbance of the individual sub-cells and reflectance of the tandem device as a function of the recombination contact layer. Either 80 nm ITO or 30 and 80 nm μ c-Si is applied in the simulation in the recombination contact between both sub-cells. Independent of the μ c-Si layer thickness, the reflectance in the wavelength range between 800 and 900 nm and in the range between 1000 and 1100 nm is reduced. Thus the absorbance and with that the EQE of the silicon sub-cell is enhanced when using μ c-Si instead of ITO. The reflection below 800 nm and with that the absorption of the perovskite is merely affected. Details of the simulation can be found in the experimental section.