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Monolithic Perovskite/Silicon Tandem Photovoltaics with Minimized Cell-to-Module Losses by Refractive-Index Engineering

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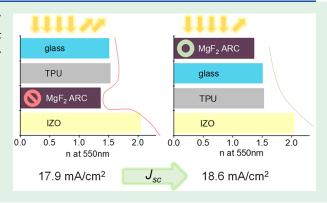
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ABSTRACT: We report 26.2% efficient monolithic perovskite/silicon tandem single-cell solar modules with a short-circuit current density of 18.6 mA/cm², enabled through enhanced optical design.



onolithic perovskite/silicon tandem solar cells have attracted significant attention in the photovoltaic (PV) community because of their high power conversion efficiency (PCE) potential (theoretical limit of ~45%; ^{1–4} reported experimental values close to 30% ^{5–7}), coupled with the relative ease of integration of perovskites with mainstream crystalline silicon (c-Si) solar cells. Although our monolithic solution-processed perovskite/silicon tandem cells exhibit PCEs > 28%, ⁸ we found that upon encapsulation, as required for true outdoor deployment, single-cell perovskite/silicon tandem mini-modules (hereafter termed modules; see Figure 1A for the structure and the Supporting Information for details) may show significant cell-to-module losses.

Our modules are fabricated by laminating the tandem cell between two glass sheets using thermoplastic polyurethane (TPU) as an encapsulant and butyl rubber as edge sealant. With this configuration, the cell-to-module PCE losses are as high as 3.2% absolute, reducing the PCE from 28.9% to 25.7% (Figure 1B). This loss is mainly due to a 1.7 mA/cm² drop in short-circuit current density ($J_{\rm sc}$) from cell (19.6 mA/cm²) to module (17.9 mA/cm²). From external quantum efficiency (EQE) and absorption measurements of the tandems (Figure 1C), we infer that this $J_{\rm sc}$ loss is caused by the introduction of glass and TPU, resulting in front reflection/escape and parasitic absorption of incident light. Here, we reduce this front reflection loss by optical redesign of the module through refractive-index engineering.

First, we identified the main source of the increased front reflection. By examining the refractive indices n (at 550 nm) of the employed optical films in the perovskite/silicon tandem module (Figure 1D), we found that the MgF₂ film, used as an antireflection coating (ARC) at the cell level, impedes at the module level photon transmission into both the perovskite and silicon subcells because of a refractive-index mismatch. Moving the MgF₂ ARC from its current position to the outer surface of the front module glass can restore the increasing refractive-index gradient of front side materials, as required for efficient light incoupling. Because the refractive index *n* varies with wavelength, we carried out optical simulations of perovskite/silicon tandem modules to verify whether moving the MgF₂ ARC effectively enhances photon absorption in the subcells over the entire wavelength range of interest (280-1200 nm). This simulation employs a new modeling methodology that solves the film conformality issue when simulating a flattened perovskite top cell on a textured silicon bottom cell, representative of our tandem solar cells, by using ray tracing in combination with the

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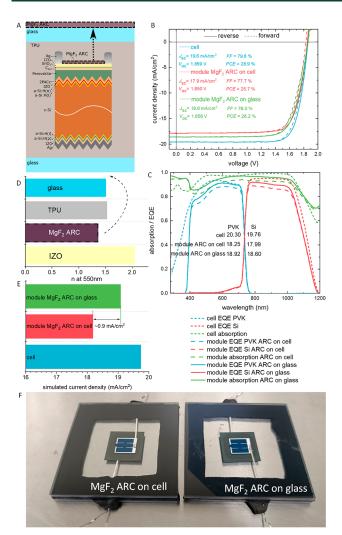


Figure 1. (A) Perovskite/silicon tandem single-cell mini-module structure and the demonstration of the optical optimization by moving the MgF₂ ARC from the cell to the top of the module front glass. (B) Experimentally measured current-density/voltage curves of a perovskite/silicon tandem cell and modules before and after optical optimization. The I-V characteristic parameters of I_{sc} opencircuit voltage (V_{oc}) , fill factor (FF), and PCE of devices are inset in the figure. The label "module MgF2 ARC on cell" refers to the tandem module with the MgF2 ARC on the top of the cell, i.e., directly encapsulating the cell without module optical optimization. The label "module MgF₂ ARC on glass" refers to the tandem module with the MgF₂ ARC on the top of the front glass, i.e., after module optical optimization. Those labels apply to other subfigures. (C) Experimentally measured EQE and absorption of a perovskite/ silicon tandem cell and modules before and after optical optimization. (D) Refractive indices n of several top films of the perovskite/silicon tandem module. The arrow indicates the optimization in this study that moving the MgF2 ARC from the cell to the top of the module front glass. (E) Simulated current densities of the perovskite/silicon tandem cell and modules. (F) Photos of experimental modules: (left) with MgF2 ARC on cell and (right) with MgF₂ ARC on glass whose left-bottom corner was taped during MgF₂ deposition so the color is different from that of other parts.

transfer matrix method. The detailed modeling method will be published elsewhere. ¹⁰ In Figure 1E, we list the simulated $J_{\rm sc}$ of (i) the bare tandem cell; (ii) the tandem module with the MgF₂ ARC on the cell, i.e., before optical module optimization; and

(iii) the tandem module with the MgF_2 ARC on the module front glass, i.e., after optimization. Note that we assume perfect carrier collection at all wavelengths for both perovskite and silicon subcells after they absorb the photons (i.e., internal quantum efficiency, IQE = 1) and ignore front metal finger shading. From Figure 1E, we find that even though the module J_{sc} is always lower than that of the bare cells, by simply moving the MgF_2 ARC from the cell to the top of the module front glass one already can significantly mitigate the cell-to-module J_{sc} loss by $\sim 0.9 \text{ mA/cm}^2$.

With guidance from the above analysis and simulation, we also applied the optimization in experiment. Figure 1F shows the experimental modules with MgF₂ ARC on the cell (left) and on the top of the module front glass (right). From the experimentally measured EQE and absorption in Figure 1C, we effectively find that by moving the MgF₂ ARC from the cell to the top of the module front glass, the front reflection is generally reduced (i.e., absorption data increases in the plot) for almost all wavelengths over 280–1200 nm, and the EQEs of perovskite and silicon subcells are both increased. As a result, as the experimental current-density/voltage (J/V) data shown in Figure 1B, the tandem J_{sc} increases by 0.7 mA/cm² (from 17.9 to 18.6 mA/cm²) in the new configuration, enabling a monolithic perovskite/silicon tandem module PCE increment from 25.7% to 26.2%.

This work highlights the need to investigate and account for cell-to-module losses in order to take full advantage of the high PCE potential of perovskite/silicon tandems.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsenergylett.2c01142.

Experimental procedures, including Si bottom cell fabrication, perovskite top cell fabrication on Si substrate, device encapsulation, solar cell characterization; short-circuit current density summary of experimental samples, with Figure S1 (PDF)

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Author Contributions

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L.X., J.L., M.D.B., M.B., and F.T. conceived ideas, designed experiments, and analyzed the data; J.L., M.B., F.T., L.X., M.D.B., W.Y., F.X., J.K., T.A., A.R., and E.A. prepared tandem devices and the related optical characterization samples; L.X. extracted related refractive index of films and did the optical simulation; J.L. and F.T. measured J-V, EQE, and absorption; L.X. wrote the paper and plotted the figures; S.D.W supervised the project.

Notes

The authors declare no competing financial interest.

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