Single-Crystal MAPbI₃ Perovskite Solar Cells Exceeding 21% Power Conversion Efficiency

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Supporting Information

ABSTRACT: Twenty-micrometer-thick single-crystal methylammonium lead triiodide (MAPbI₃) perovskite (as an absorber layer) grown on a charge-selective contact using a solution space-limited inverse-temperature crystal growth method yields solar cells with power conversion efficiencies reaching 21.09% and fill factors of up to 84.3%. These devices set a new record for perovskite single-crystal solar cells and open an avenue for achieving high fill factors in perovskite solar cells.

In the past few years alone, hybrid metal halide perovskite materials have revolutionized the field of low-temperature-processed solar cells, providing devices with power conversion efficiencies (PCEs) that compete with the most established decades-old commercial photovoltaic technologies. To date, studies on perovskite solar cells (PSCs) have principally been on polycrystalline film PSCs (Pc-PSCs). The record efficiency for Pc-PSCs, currently at 24.2% PCE, is still far from their theoretical Shockley–Queisser limit (SQL), which is ~30.5% PCE for a single-junction cell based on methylammonium lead triiodide (MAPbI₃). To approach the SQL, it is essential to improve the device’s fill factor (FF) (FF ≈ 90% in the SQL for MAPbI₃ PSCs), which contributes to the overall PCE. The FF is perhaps the most challenging figure of merit to improve for Pc-PSCs because losses in FF are mainly governed by the ideality factor that is related to nonradiative bulk and interface carrier recombination. Despite marked efforts in improving the FF, polycrystalline thin films have significant parasitic nonradiative carrier recombination due to their inherent grain size and surface defects.

In theory, single-crystal perovskites, with their orders of magnitude lower defect density and higher carrier diffusion lengths compared to their polycrystalline counterparts, offer a chance for PSC technology to overcome the limitation of polycrystalline thin films and get as close as practical to the SQL. Unfortunately, because of the challenges with their thickness control and typically device-incompatible solution-growth conditions, only a handful of groups have led the charge in the development of single-crystal-based PSCs (SC-PSCs). SC-PSCs reached their highest (PCE of 17.8% with FF of 78.6%) in a 2017 report by Huang and co-workers. Here, we realize highly efficient SC-PSCs with PCEs reaching 21.09% and FFs of up to 84.3% (under 1 sun illumination AM 1.5G). These devices, based on a ~20 μm thick MAPbI₃ single-crystal absorber layer in an inverted p-i-n architecture, set a new record for SC-PSC PCE and a new benchmark for potential FFs that PSCs should aim for, which Pc-PSCs have struggled to achieve.

The crystals were grown using a simple solution-space-limited inverse-temperature crystal growth method (see the Supporting Information). The crystal structure and optical absorption edge of these crystals are that of typical MAPbI₃ single crystals (see Figures S1 and S2). A scanning electron microscopy (SEM) cross-sectional image of the MAPbI₃ SC-PSC (Figure 1a) shows that the active area is pinhole- and grain boundary-free (see also top-view SEM image of the crystal, Figure S3). The smooth surface allows for the full coverage of the charge transport layers, preventing direct crystal-to-metal-electrode contact. The device follows an ITO/poly(triarylamine) (PTAA)/MAPbI₃ single-crystal/C₆₀/bathocuproine (BCP)/copper (Cu) architecture with the corresponding energy band diagram illustrated in Figure 1b. The crystal thickness in devices is typically around 20 μm (see distribution in Figure S4).

Figure 1c shows the current–voltage characteristics of our best performing cell. The reverse-scan short-circuit current density (Jsc), open-circuit voltage (Voc), and FF are 23.46 mA cm⁻², 1.076 V, and 83.5%, respectively, yielding a PCE of...
21.09% with negligible hysteresis. The corresponding external quantum efficiency (EQE) and integrated $J_{SC}$ are shown in Figure 1d. To exclude the effect of any potential error in aperture area determination, the $J_{SC}$ of the champion device was verified and corrected from the integrated current calculated from the EQE. The calculated $J_{SC}$ from the EQE measurement was only 1.2% lower than that obtained by an $I–V$ scan for the device, which is within the expected error of solar cell $J–V$ measurements. The steady-state maximum power output (SPO) at 0.93 V bias for the champion cell (Figure 1e) shows that the PCE reaches over 21%, which sets a new efficiency benchmark for SC-PSCs. Our best fabricated MAPbI$_3$ SC-PSCs with PCEs reaching 21.09%, which sets a new efficiency benchmark for SC-PSCs. While there is still room for larger-area SC-PSCs and substantial interfacial optimization, the high PCE and FF highlight the promise of single crystals for advancing perovskite device technology, which could be a parallel development path to the one taken by their polycrystalline counterparts.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsenergylett.9b00847.

Details of device fabrication and characterization, figures as described in the text, and TOC abbreviations (PDF)

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**Notes**

The authors declare no competing financial interest.

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