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Short Communication

26 mA cm^{-2} J_{SC} achieved in the integrated solar cells

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Organometal halide perovskite has drawn much attention due to their high light-absorption coefficient, outstanding carrier mobility, and long-range charge-transport lengths. The most remarkable progress made by this type of perovskite materials is in the field of photovoltaics [1–4]. The power conversion efficiency (PCE) of perovskite solar cells (PSCs) has exceeded 25% since they were first used as the active layer in solar cells in 2009 [5], which is comparable to single-crystal silicon solar cells and approaching the Shockley–Queisser efficiency limit of single-junction cells [6]. To date, high-performance PSCs are dominated by the typical ABX_3-type perovskite materials with a bandgap (E_g) in the range of 1.5–1.6 eV, resulting in light absorption spectra with edges mostly below 850 nm. The lack of light-harvesting in the near-infrared (NIR) region could limit the further improvement of the efficiency [7,8]. To this end, some PSCs were fabricated by incorporating Sn or Sn/Pb alloy into perovskite, leading to reduced bandgaps and extended photoresponse. However, these Sn-based PSCs not only show inferior PCEs but also suffer from poor stability. Other narrow-E_g PSCs were made via the tandem strategy, where complementary absorption spectra by different subcells lead to broadened overall absorption. Nevertheless, the tandem structure increases the difficulty of device fabrication.

An effective alternative strategy is to combine and integrate bulk heterojunctions (BHJ) organic solar cells (OSCs) and PSCs, here named integrated solar cell (ISC) [9]. Unlike tandem devices, the OSC materials can be directly deposited onto the perovskite layer by spin-coating using orthogonal solvents. Accordingly, the fabrication of ISCs is simple and cost-effective since there is no tunnel junction in series-connected tandem solar cells. The first ISC was reported by Zuo and Ding (online October 2014) [10], where an ISC with a PCE of 8.80% and a J_{SC} of 13.93 mA cm^{-2} were achieved. Recent studies have demonstrated that preparing an NIR BHJ layer onto the perovskite layer can effectively broaden the absorption and increase the J_{SC} of devices without sacrificing the V_{OC} of PSCs. For example, Liu et al. [11] succeeded in harvesting the NIR photons by using a narrow-E_g polymer donor (PBDTT-SeDPP) that enabled an efficiency of 12.0% along with a J_{SC} of 20.6 mA cm^{-2}. Tan and co-workers [12] used a narrow-E_g nonfullerene acceptor (IEICO) and achieved much higher efficiency (14.3%) and J_{SC} (21.2 mA cm^{-2}) in ISCs. As shown by these studies, despite that the absorption profiles were (in some cases) extended to the NIR region, but the J_{SC} was limited to around 20 mA cm^{-2} and the external quantum efficiency (EQE) of the ISC is still low in the NIR region.

Herein, we demonstrate a facile approach to broaden the photoresponse and increase the efficiency of PSCs by depositing a ternary organic blend [13,14] that consists of a new wide-E_g polymer (S2), a narrow-E_g small molecule (Y6) [15] and $PC_{61}BM$ onto the perovskite layer. Benefiting from the good band alignment, efficient charge transfer between the BHJ and perovskite is realized. Compared to the control device without the BHJ layer, the ISC with the ternary BHJ layer exhibits an extended EQE spectra approaching 1000 nm, which is much redder than the perovskite layer alone. Notably, the highest EQE value approaches 90% in the NIR region. As a result, a high J_{SC} of 26.7 mA cm^{-2} is achieved by our ISC, which is significantly higher than the neat PSCs (20.16 mA cm^{-2}). To the best of our knowledge, this is the champion value among all ISCs. Our approach can be readily expanded to other classes of perovskite active layers to increase the NIR absorption and thus the efficiency of PSCs.

The chemical structures and absorption spectra of the photovoltaic materials used in ISC are shown in Fig. 1a and c, respectively. The ternary organic blend consists of one new polymer donor named S2 synthesized following our previous method (see the Supplementary materials) [16], and two small-molecule acceptors Y6 and $PC_{61}BM$. The absorption edges of the perovskite and BHJ films are 800 and 900 nm, respectively. When integrated, the absorption spectrum of the perovskite/BHJ film afforded by the dual active layer covers the whole visible region and partial NIR

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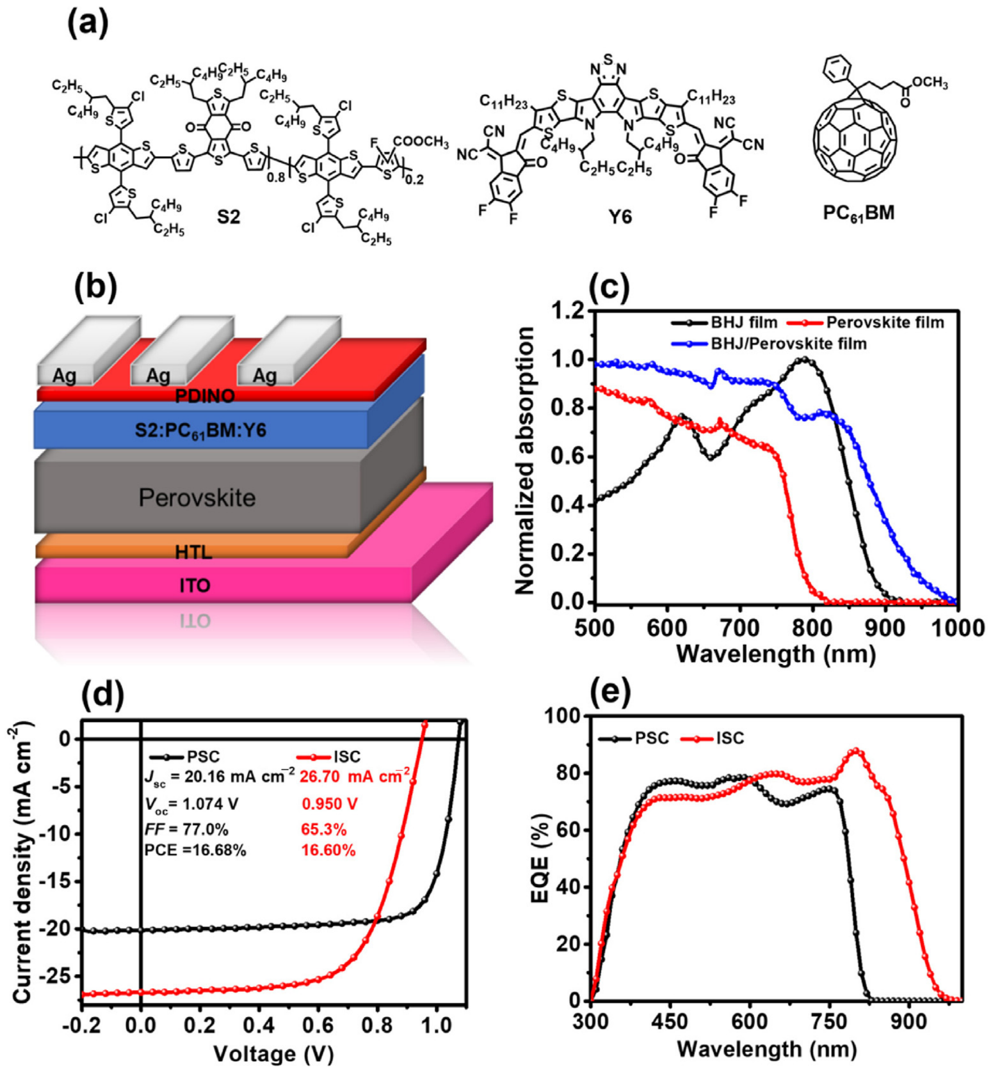

Fig. 1. A record J_{SC} of 26.7 mA cm^{-2} is achieved from integrated organic bulk heterojunction/perovskite solar cells. (a) Chemical structure of photovoltaic materials in ternary BHJ films; (b) Device structure of the ISC; (c) Normalized absorption spectra of BHJ, perovskite and BHJ/perovskite films; (d) *J-V* characteristics of two devices; (e) EQE spectra of the PSC and ISC.

region. Notably, compared to BHJ film, an obvious red-shift was observed in BHJ/perovskite film most likely due to the change in molecular ordering of BHJ film. In the ISC, the perovskite underlayer may have an effect on molecular ordering of BHJ layer above. Some of previous works have shown that films could show an expanded absorption when *J*-aggregation in blend films were formed [14,17]. In our case, perovskite film may contribute to the broadening of BHJ blend film spectrum from 900 to 100 nm by tuning molecular ordering and orientation of BHJ, leading to an enhanced J_{SC} in the ISC. We note that the hypothesis is pending for further validation and there is no direct evidence yet. More characterization on the morphology for ISC is ongoing.

Next, ISCs with a structure of ITO/P3CT-Na/$(FA_{0.17}MA_{0.94}PbI_{3.01})_{0.95}(PbCl_2)_{0.05}$/(S2:PC$_{61}$BM:Y6)/PDINO/Ag are designed and fabricated as shown in Fig. 1b, where ternary organic BHJ is deposited directly on top of the perovskite layer via spin-coating. In comparison, the control PSCs with a structure of ITO/P3CT-Na/$(FA_{0.17}MA_{0.94}PbI_{3.01})_{0.95}(PbCl_2)_{0.05}$/PC$_{61}$BM/PDINO/Ag are also fabricated, where only PC$_{61}$BM is used as the electron transport layer. The *J-V* curves of the corresponding devices are shown in Fig. 1d and S1 (online). The control PSC yields a PCE of 16.68% with a V_{OC} of 1.07 V, a J_{SC} of 20.16 mA cm^{-2} and a fill factor (*FF*) of 77.0%. In order to further improve J_{SC} and NIR region photoresponse, we prepared ISCs with organic ternary BHJ (weight ratio: S2:PC$_{61}$BM:Y6 = 1:1:2). Although the two types of devices show comparable PCEs, the J_{SC}s, from the perovskite to BHJ/perovskite-based cells, sharply increase from 20.16 to a remarkable 26.70 mA cm^{-2}. Compared to the control PSC, the addition of the BHJ enhances the J_{SC} by 6.54 mA cm^{-2}. To the best of our knowledge, 26.70 mA cm^{-2} is the highest J_{SC} value reported so far for integrated ISCs. In addition, the neat BHJ cell was also fabricated using the conditions for making the best ISC (see the Supplementary materials for details). Fig. 1e demonstrates the EQE curves of the PSC and ISC. The PSC shows a broad plateau with photoresponse below 80% in the visible range (300–800 nm) but a cutoff at 760 nm. In sharp contrast, the ISC demonstrates a nearly 90% EQE at 800 nm and an averaged EQE over 70% in the NIR region of 780–880 nm, indicating efficient charge separation and transfer between BHJ and perovskite. The integrated J_{SC} from EQE for PSC and ISC are 19.67 and 25.57 mA cm^{-2}, respectively, which are in good agreement with the values derived from the *J-V* curves (within 4% error). The high EQE in NIR region is unprecedented in integrated ISCs and could be mainly attributed to the effective charge separation and low energy losses in the OSC based on Y6. However, the *FF* for the ISC is lower than that of PSC, possibly due to large contact resistance at the BHJ/perovskite interface. More optimization is ongoing to further improve the device performance.

In conclusion, a high J_{SC} of over 26 mA cm^{-2} is achieved in single-junction medium-E_g PSC by integrating a BHJ ternary organic layer into the PSC. The EQE of the integrated ISC extends to 950 nm and the highest value is ~90% at 800 nm. Our results demonstrate that the perovskite/BHJ system has huge potential in breaking the single-junction S-Q efficiency limit.

Conflict of interest

The authors declare that they have no conflict of interest.

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Author contributions

Xugang Guo proposed the research and directed the study. Huiliang Sun carried out the materials synthesis. Huiliang Sun, Qiaogan Liao and Bolin Li performed cell fabrication and characterizations. Huiliang Sun, Qiaogan Liao and Xugang Guo analyzed the data and prepared the manuscript. Huiliang Sun and Qiaogan Liao contributed equally to this work.

Appendix A. Supplementary materials

Supplementary materials to this article can be found online at https://doi.org/10.1016/j.scib.2019.10.005.

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