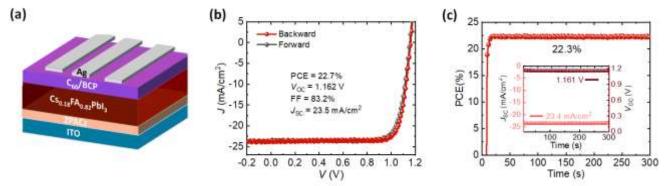
## Simultaneous Interfacial and Grain-Boundary Passivation for Highly Efficient Inverted Methylammonium-Free Perovskite Solar Cells

## Saba Gharibzadeh

Paul Fassl,<sup>a,b</sup> Ihteaz M. Hossain,<sup>a,b</sup> Pascal Rohrbeck,<sup>c</sup> Markus Frericks,<sup>d,e</sup> Moritz Schmidt,<sup>a,b,f</sup> The Duong,<sup>g</sup> Motiur Rahman Khan,<sup>a</sup> Tobias Abzieher,<sup>a</sup> Bahram Abdollahi Nejand,<sup>a,b</sup> Fabian Schackmar,<sup>a,b</sup> Osbel Almora,<sup>h</sup> Thomas Feeney,<sup>a</sup> Roja Singh,<sup>a,b</sup> Uli Lemmer,<sup>a</sup> Jan P. Hofmann,<sup>d,e</sup> Stefan A.L. Weber,<sup>c</sup> and Ulrich W. Paetzold<sup>a,b</sup>

- <sup>a</sup> Light Technology Institute, Karlsruhe Institute of Technology, Engesserstrasse 13, 76131 Karlsruhe, Germany.
- <sup>b</sup> Institute of Microstructure Technology, Karlsruhe Institute of Technology, Hermann-von Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany.
- <sup>c</sup> Max Planck Institute for Polymer Research, Department of Physics at Interfaces, Ackermannweg 10, 55128 Mainz, Germany.
- <sup>d</sup> Technical University of Darmstadt, Department of Materials and Earth Sciences, Surface Science Laboratory, Otto-Berndt-Strasse 3, 64287 Darmstadt, Germany.
- <sup>e</sup> InnovationLab GmbH, Speyerer Strasse 4, 69115 Heidelberg, Germany.
- <sup>f</sup> Center for Nanophotonics, AMOLF, 1098 XG Amsterdam, The Netherlands
- g School of Engineering, The Australian National University, Canberra, 2601 Australia.
- <sup>h</sup> Institute of Advanced Materials, Universitat Jaume I, 12006 Castelló, Spain saba.gharibzadeh@kit.edu

Inverted p-i-n perovskite solar cells (PSCs) have proven their potential for fabricating highperformance perovskite-based tandem photovoltaics [1]. However, severe non-radiative recombination at the perovskite/electron transport layer and at the grain boundaries (GBs) still limits their open-circuit voltage (V<sub>OC</sub>) and fill factor (FF) as compared to their n-i-p counterparts. We introduce a novel dual passivation approach using phenethylammonium chloride (PEACI) to simultaneously passivate the GBs and the perovskite/C60 interface by using PEACI:PbCl2 as the additive and PEACI for surface treatment, respectively. Thereby, we achieve a substantial enhancement in charge carrier lifetime and quasi-Fermi level splitting compared to reference devices. Thereby, we achieve a substantial enhancement in charge carrier lifetime and quasi-Fermi level splitting compared to either of the individual passivation strategies or reference devices. By analyzing cathodoluminescence, scanning electron microscopy, X-ray/ultraviolet photoelectron spectroscopy and Kelvin probe force microscopy measurements, we attribute the positive effects to the formation of a heterogeneous 2D Ruddlesden-Popper perovskite at the film surface and GBs, which leads to efficient chemical passivation of GB and surface defects as well as additional electronic passivation at the perovskite/C<sub>60</sub> interface. This results in one of the highest reported PCEs for p-i-n PSCs of 22.7% with a remarkable V<sub>OC</sub> and FF of 1.162 V and 83.2%, respectively. Our study researches the advanced use of dual passivation as key mechanism to manage the detrimental defects both in the GBs and the surface of the perovskite layer to achieve highly efficient p-i-n PSCs. The work paves the way for the development of high-efficiency two-terminal perovskite-based tandems photovoltaics.



**Figure 1:** (a) Schematic of the employed perovskite solar cell configuration with a layer stack sequence of ITO/2PACz/perovskite/ $C_{60}$ /BCP/Ag. (b) *J-V* characteristics and (c) maximum power point (MPP) tracking of the best-performing dual passivation perovskite solar cell, demonstrating a stabilized PCE of 22.3%. The inset shows the stabilized  $J_{SC}$  and  $V_{OC}$ .

## REFERENCES

[1] Zheng, X., Hou, Y., Bao, C., Yin, J., Yuan, F., Huang, Z., Song, K., Liu, J., Troughton, J., Gasparini, N. and Zhou, C, Nature Energy, 2020, 5(2), pp.131-140.