Improving Perovskite Solar Cells: Towards a Device Model

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To improve the performance of existing perovskite solar cells (PSCs), a detailed understanding of the underlying device physics during their operation is essential.

As a first step, we have developed and validated a device model[1] that describes the operation of PSCs and quantitatively explains the role of contacts and of (doped) transport layers, carrier generation, drift and diffusion of carriers and recombination. We fit the simulation to experimental data of vacuum deposited $CH_3NH_3PbI_3$ solar cells over multiple thicknesses. By doing so, we identify a unique set of parameters and physical processes that describe these solar cells. Recombination at material interfaces (HTL/perovskite and perovskite/ETL) is the dominant loss channel limiting the device performance and passivation of these interfaces increases the power conversion efficiency (PCE) of these devices by 40%. Finally, we issue guidelines to increase performance and show that a PCE beyond 25% is within reach.

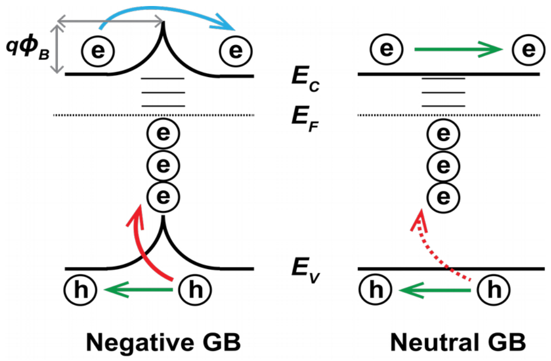

Figure 1. *Charge carrier movement through a negative and a neutral grain boundary. In the case of a negative grain boundary, electron transport is weakened due to the potential barrier ($q\varphi_B$) and non-radiative recombination between holes and trapped electrons is strong. For a neutral ground boundary, electron transport is relatively unaffected, and non-radiative recombination is weak.*

Grain boundaries are ubiquitous in polycrystalline films and are studied extensively in CIGS, poly-Si and CdTe solar cells. The Seto model[2] is able to successfully describe the grain boundary physics in these doped solar cells, see Fig. 1. However, PSCs are different. Perovskites are lightly doped materials and due to the presence of ionic defects it is likely that the grain boundaries are charged when empty and neutral when filled, in contrast to the basis of the classic Seto model. Therefore, a different perspective on defect physics is essential for PSCs. We include grain boundaries in our model (see Fig. 2) and fit the simulation to vacuum deposited $CH_3NH_3PbI_3$ solar cells in p-i-n and n-i-p configuration.[3]

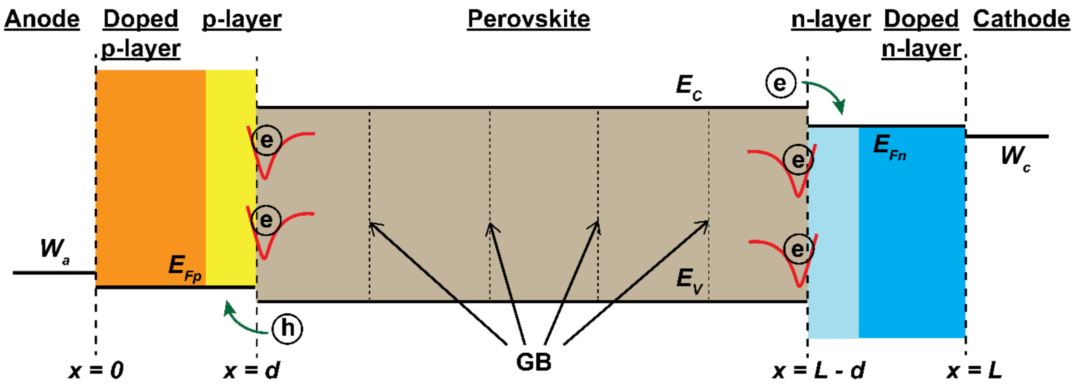

Figure 2. *The p-i-n device skeleton showing the energy levels, interface electron traps (red) and grain boundaries (dashed lines). Upon illumination, free electrons and holes are transported through the respective materials and are extracted at the electrodes.*

Our model quantitatively explains (for both p-i-n & n-i-p cells) the light intensity dependence of the open-circuit voltage and fill-factor, delineating the recombination dynamics at grain boundaries and interfaces under different operating conditions. We find that despite the presence of traps at grain boundaries, their neutral (when filled) disposition along with the long-lived nature of holes leads to the high-performance of PSCs. We also give an estimate of the defect ion density in these solar cells. Furthermore, we shed light on the role of charged grain boundaries which may exist under some conditions (under/over stoichiometric preparation).

[1] T.S. Sherkar, C. Momblona, L. Gil-Escrig, H.J. Bolink, and L.J.A. Koster, Adv. Energy Mater. 1602432 (2017).
[2] J. Y. Seto, J. Appl. Phys. **1975**, *46*, 5247-5254.
[3] T.S. Sherkar, C. Momblona, L. Gil-Escrig, J. Ávila, M. Sessolo, H. Bolink, and L.J.A. Koster**,** ACS Energy Lett. **2**, 1214 (2017).