

Operando spectroscopic imaging of the solar cell energy landscape

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Metal halide perovskites (MHP) feature essential properties, such as high absorption coefficients, low exciton binding energy, long charge carrier diffusion lengths, and balanced charge transport, that make them ideal candidates for high-efficiency, low-cost photovoltaics. However, the interface between the perovskite active layer and charge transport layers (CTLs) plays a crucial role in determining both efficiency and stability. This work aims to develop a novel strategy to better understand and optimize these interfaces. To achieve this, we integrate Photoluminescence (PL) and Electron Beam Induced Current (EBIC) studies under operando conditions, allowing us to correlate electronic and optoelectronic properties at the MHP/CTL interface. Our investigation focuses on lateral heterojunction (LHJ) devices, which feature carrier-selective materials for back contacts and allow direct access to the active layer from the top [1, 2]. Specifically, we studied LHJ configurations using NiOx and TiO₂ as selective contacts, interacting with double-cation Cs_{0.3}FA_{0.7}Pb(Br_{0.1}IO_{0.9})₃ and triple-cation Cs_{0.05}(MA_{0.17}FA_{0.83})_{0.95}Pb(Br_{0.17}IO_{0.83})₃ perovskites. To effectively implement these techniques under operando conditions, a dedicated measurement protocol has been designed. Surface illumination (LEDs for PL, electron beam for EBIC) is applied with a specific focus on the MHP/CTL interfaces. The measurements follow a structured sequence of applied voltages, including polarity switching, to simulate operational conditions. In particular, measurements cycles between 0 and +V and 0, +V, 0, -V will be tested. These sequences are designed to identify and distinguish between different contributing effects, helping to highlight features and limitations of the device structure. During these experiments, the evolution of the active layer at the interfaces is continuously monitored, with both current and time being tracked. Relaxation periods under dark and 0 V bias are introduced between voltage steps to assess the potential for recovery. This comprehensive approach enables the correlation of dynamic interfacial changes with operating conditions, offering deeper insight into interface behaviour and its impact on device performances. To validate this method, PL mapping with the application of an electrical bias have been carried out, to investigate the interfacial evolution under various voltage conditions in a non-controlled environment (i.e., in air) and under controlled nitrogen atmosphere (i.e., in a cryostat). We thus record microscopically resolved absolute PL spectra of the perovskite on top of the contacts and along the channel to assess bandgap variations and calculate the local quasi-Fermi level splitting ($\Delta\mu$). We find that the photocurrent increases when applying voltage, which means functional extraction under bias. Moreover, from the maps it is possible to observe a quenching near one of the two interfaces, indicating good carrier extraction. Looking at the spectrum, a homogeneous band gap shift across the sample appears, as an effect of light soaking and electric field.

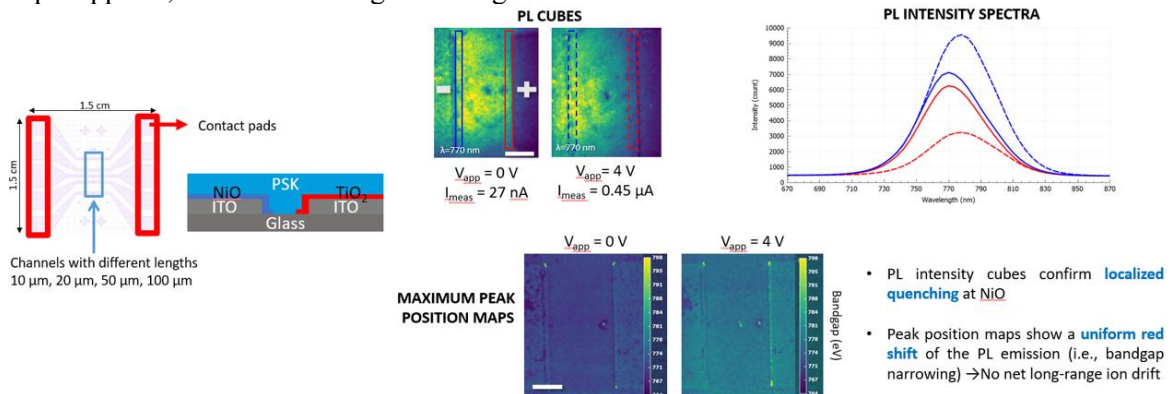


Figure 1. Schematic of the lateral heterojunction device and first results.

References

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