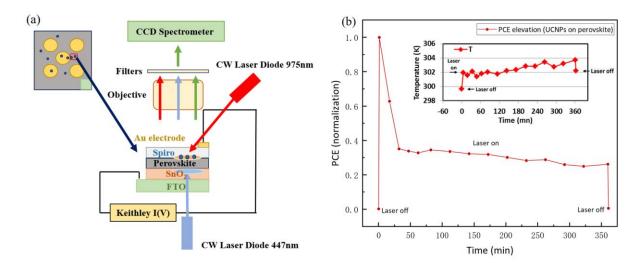
## In-situ Temperature Monitoring of Perovskite Solar Cells Degraded under Illumination

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Perovskite solar cells (PSCs) are considered to be one of the most promising emerging photovoltaic technologies due to their high power conversion efficiency (PCE) achievable by near-room-temperature solution-processing. In terms of PCE, this technology has witnessed a rapid growth from  $\sim 3$  % since early experiments in 2009 to > 26% nowadays. Nevertheless, the material and device instability of PSCs is still to be resolved, as they tent to degrade when exposed to normal environmental factors during solar cell functioning, such as oxygen, humidity, and light. Currently, on-going international research efforts have been focused on the investigations to understand the degradation mechanisms and to propose engineering methods to mitigate such effects.

Under this context, in this work, we applied local electro-optical scanning means to monitor in-situ the thermal property change of triple-cation PSCs at the nanoscale during degradation. The present scanning method relies on the nano-temperature probes based on inorganic Er<sup>3+</sup> doped NaYF<sub>4</sub> colloidal nanoparticles capable to perform photon upconversion (below termed as UCNPs). These UCNPs can be excited by near-infrared photons which are transparent to the PSCs and emit multiple well-defined upconversion fluorescence peaks at different wavelengths intrinsic to the multiphoton absorption and energy transfer processes. In particular, the peak ratio between the two upconversion fluorescence peaks in the visible spectrum ( $\lambda_{510-532nm}/\lambda_{532-565nm}$ ) has been shown to be highly sensitive to the local temperature in their vicinity.<sup>1-2</sup> Herein, we inserted purposefully UCNPs on the different interfaces of functional PSCs based on triple-cation Cs<sub>0.05</sub>(FA<sub>0.83</sub>MA<sub>0.17</sub>)<sub>0.95</sub>Pb(I<sub>0.83</sub>Br<sub>0.17</sub>)<sub>3</sub>. The surfaces/interfaces under study include the one situates on top of the gold back contact electrode, in between the HTL/back contact, and between the HTL/perovskite interfaces. Figure 1(a) exhibits the schematic of our experimental setup. During a typical in-situ experiment, the device under study was exposed continuously to the illumination of a blue laser ( $\lambda = 447$  nm) in air which serve as the degradation sources. Over the degradation period, both the local temperature evolution at the abovementioned surfaces/interfaces and the photovoltaic parameters were monitored. As shown in Figure 1(b), in the configuration where the UCNP temperature probes were inserted at the perovskite/HTL interface, we first observed a quasi-instantaneous (within  $\sim 2s$ ) temperature elevation of 2°C upon turning on the blue laser. After this initial stage, the interfacial temperature of the device continuously increases accompanied with a progressive decrease of PCE which should originate from device degradation. In this communication, we will discuss the different phenomena observed and in particular the relationship between the temperature elevation and the PCE decrease.



**Figure 1**(a) Schematic diagram of our experimental setup; (b) The observed PCE and temperature evolution of the PSCs under degradation (with UCNP temperature probes inserted at the HTL/perovskite interface).

## References

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(2) M.S. Sebag et al., Microscopic Evidence of Upconversion-Induced Near-Infrared Light Harvest in Hybrid Perovskite Solar Cells. *ACS Appl. Energy Mater.* 1, 3537-3543 (2018)