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**From Morphology to Interfaces to Tandem Geometries: Enhancing the Performance of Perovskite/Polymer Solar Cells<sup>1</sup>**

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We have taken a new approach to develop mesoporous lead iodide scaffolds, using the nucleation and growth of lead iodide crystallites in a wet film. A simple time-dependent growth control enabled the manipulation of the mesoporous lead iodide layer quality in a continuous manner. The morphology of lead iodide is shown to influence the subsequent crystallization of methylammoniumleadiodide film by using angle-dependent grazing incidence x-ray scattering. The morphology of lead iodide film can be fine-tuned, and thus the methylammoniumleadiodide film quality can be effectively controlled, leading to an optimization of the perovskite active layer. Using this strategy, perovskite solar cells with inverted PHJ structure showed a *PCE* of 15.7 per cent with little hysteresis. Interface engineering is critical for achieving efficient solar cells, yet a comprehensive understanding of the interface between metal electrode and electron transport layer (ETL) is lacking. A significant power conversion efficiency (PCE) improvement of fullerene/perovskite planar heterojunction solar cells was achieved by inserting a fulleropyrrolidine interlayer between the silver electrode and electron transport layer. The interlayer was found to enhance recombination resistance, increases electron extraction rate and prolongs free carrier lifetime. We also uncovered a facile solution-based fabrication of high performance tandem perovskite/polymer solar cells where the front sub-cell consists of perovskite and the back sub-cell is a polymer-based layer. A record maximum PCE of 15.96 per cent was achieved, demonstrating the synergy between the perovskite and semiconducting polymers. This design balances the absorption of the perovskite and the polymer, eliminates the adverse impact of thermal annealing during perovskite fabrication, and affords devices with no hysteresis.

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