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Ionic liquids tailoring crystal orientation and electronic properties for stable and high fill factor perovskite solar cells

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The crystallization behavior of metal halide perovskite thin films has a profound influence on the resulting defect densities, charge carrier properties and photovoltaic performance characteristics. Herein, we introduce a pyrrolidinium-based ionic compound (Pyr14BF4) into the perovskite to investigate the impact of ionic liquids on the crystal growth of perovskite films. Using time-resolved measurements, we probe the charge generation, transport and recombination behavior in these films and related devices. We find that ionic liquids can tailor the crystal growth from a disordered to a preferential corner-up orientation during film formation and accordingly increase the charge carrier mobility to accelerate electron transport and extraction. The highest power conversion efficiency achieved based on ionic liquid-containing devices is up to 21.49% and is accompanied by a high fill factor of 0.87. Via operando grazing-incidence small- and wide-angle X-ray scattering measurements, we observe a light-induced lattice compression and grain fragmentation in the control devices, whereas the ionic liquid-containing devices exhibit a slight light-induced crystal reconstitution and stronger tolerance against illumination. Under ambient conditions, the non-encapsulated Pyr14BF4-containing device maintains 97 % of its initial efficiency after 4368 h. Our results reveal the crucial role of ionic liquids in perovskite crystallization, charge carrier kinetics and device stability.

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