Probing the nanoscale degradation mechanisms of perovskite photovoltaic materials with advanced atomic force microscopy

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Perovskite photovoltaics offer a promising approach to flexible, lightweight, and printable solar cells with high efficiencies, however their limited operational lifetime is the major barrier to commercialisation. Perovskite devices have complex micro/nano-structures and multi-layered architectures, which respond to a variety of relevant stress conditions: heat, visible light, UV, oxygen, water, electrical bias. Separating and elucidating the mechanisms of the various degradation effects and interplays presents a key measurement challenge for the progress of perovskite photovoltaics.

Advanced modes of atomic force microscopy (AFM) provide access to nanoscale characterisation of structural, functional, and dynamic properties of thin film electronic materials. In this contribution we will make use of AFM modes including (photo)-conductive AFM, Kelvin probe force microscopy, pump-probe electrostatic force microscopy, and tip-enhanced photoluminescence spectroscopy. These measurement modes provide complementary insights into the nanoscale degradation mechanisms of organic-inorganic lead-halide perovskites.

Considering perovskite thin films under a variety of operational stress conditions we present results showing the rapid formation of surface nanograins, which we chemically identify as PbI_2. Studies of the optical, environmental and electrical stress conditions related to nanograin formation leads to a proposed mechanism for this early-stage degradation based on ion-migration through the perovskite film. Further insight into the migration electronic and ionic charge carriers through the perovskite film is gained through microsecond time-resolved surface potential measurements along with tip-enhanced photoluminescence spectroscopy to reveal the nanoscale energetic landscape of the granular perovskite surface. The late-stage progression of perovskite film degradation in 3 spatial dimensions is assessed using cross-sectional AFM and spectroscopic studies of the sample prepared with a focussed ion beam (FIB).

These advanced AFM modes provide direct insight into the nanoscale structure-function relationships that will guide progress towards increased performance and lifetime of perovskite photovoltaics.