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In-situ Transmission Electron Microscopy Study of the Impact of External Electric Field in 2D Perovskites

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Organic-inorganic hybrid perovskites are promising candidates for next-generation solution-processed optoelectronics, but their outstanding properties are often overshadowed by their lack of stability under external environmental or electrical exposure. 2D perovskites [1-4] are an attractive alternative as they directly address the critical issue of stability of 3D perovskites. In the past few years, 2D perovskite devices have demonstrated high optoelectronics performances and an improved stability compared to their 3D counterparts [5,6]. Their unique layered structural design as shown in Figure 1 enables the use of organic spacer cations to tune their physical and chemical properties [1,3].

The operational stability of lower-dimensional perovskites has been found to be superior to that of 3D perovskites. Wang et.al. [7] discovered that at a current density of 10mAcm^{-2}, 3D perovskite declined to half of its EQE value in about 1 minute, whereas lower dimension perovskite lasted two hours under the same conditions. Yet, the stability of LEDs produced from 2D perovskite lasts for tens of hours which is significantly less than the commercial use requirement of >10000 hours (T_{50}, L_0=100 cd m^{-2})[8]. Tian et. al. [9] reported one of the most stable red quasi-2D perovskite LED in which the EL intensity was practically steady for 4 hours. Similarly, green LEDs have the longest stated lifetime in hours, whereas blue LEDs have the longest claimed lifetime in minutes [10,11]. The origin of the degradation under operation is not yet fully understood.

Using an *operando* scanning and transmission electron microscope (SEM and TEM), we investigate the effect of an external electric field applied on the 2D perovskite film $(PEA)_2PbBr_4$ where PEA i.e., phenylethyl ammonium is the organic spacer. We primarily focus on the relation between the microstructure, the electrical, and optical performance.
For this study, 2D perovskite $(PEA)_2PbBr_4$ was spin-coated on interdigitated electrodes for optical experiments and Si/SiN with platinum electrodes for TEM experiments, both allowing the monitoring of the electric field-induced effect *operando*. The evolution of the microstructure is followed using selected area electron diffraction (SAED) patterns in TEM. The evolution of the optical properties using time-resolved PL and the electrical properties by recording IV curve at different points in time.

Figure 2a shows the TEM image of the sample. Figure 2b illustrates a time-series of SAED patterns taken from the 2D perovskite $(PEA)_2PbBr_4$ thin film under an external electric field. The snapshots of SAED pattern initially and at different point in time is compared to study the impact of the electric field on the structure. The electron microscopy results are further compared with optical measurements. Different hypothesis for the structural changes i.e. phase changes [12], ion displacement [13][14], joule heating [15][16] are analysed. The findings pave the way to a better understanding into the relationship between the microstructural changes and the optical properties during external electric field application needed to develop the next generation of low energy LEDs.

a) b)

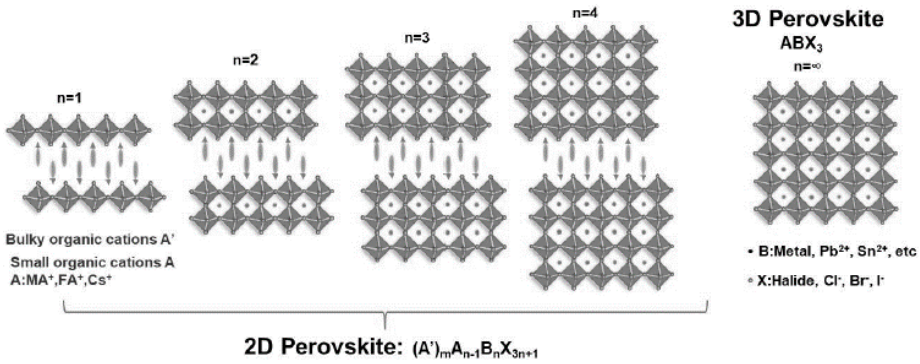
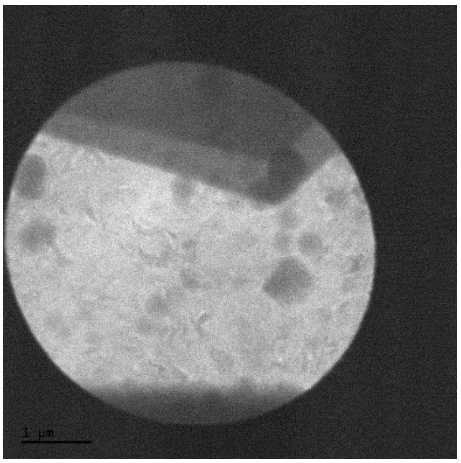
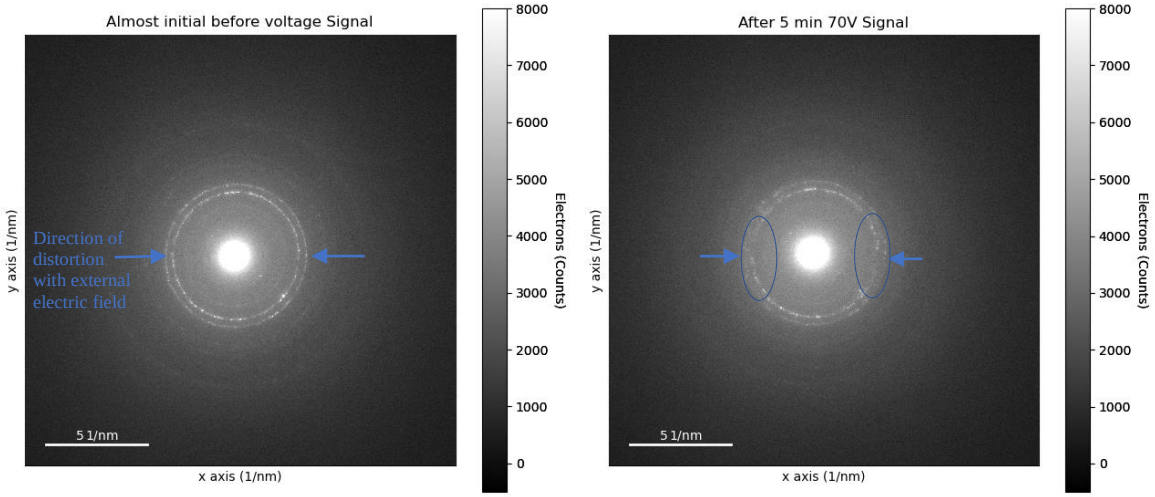


Figure 1. 2D perovskite with organic spacers. b) 3D perovskites.

a)



b)



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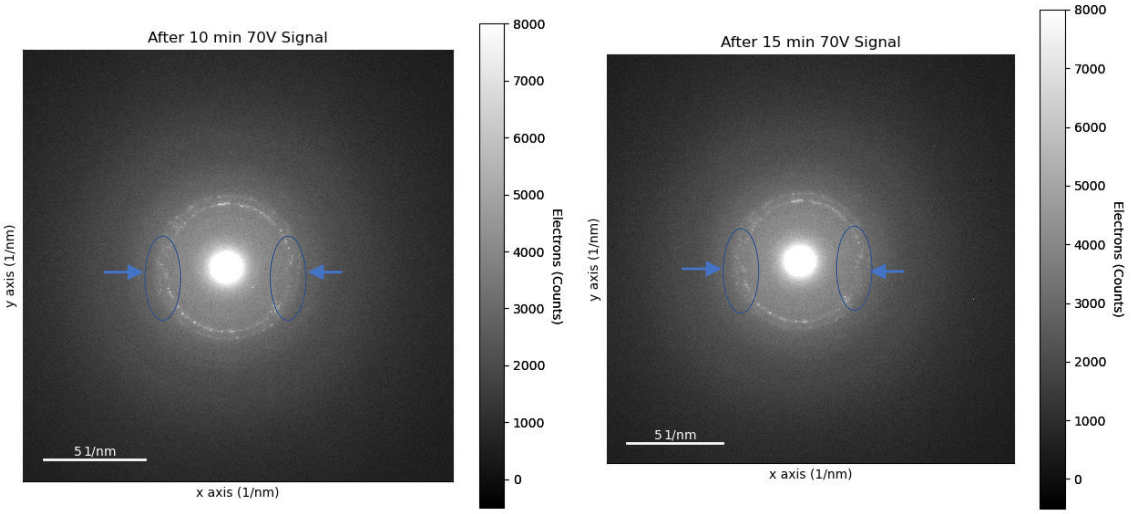

Figure 2. a) TEM image of a *2D perovskite* $(PEA)_2PbBr_4$. b) Evolution of selected area diffraction pattern with external electric field during in-situ TEM study with external electric field.

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