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Effect of Deuteration on Morphology of 2D Perovskite $(CH_3NH_3)_2Pb(SCN)_2I_2$

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The newly emerged family of organic-inorganic halide perovskites not only revolutionizes the field of photovoltaic research with an average PCE > 20%; but also triggers plentiful studies on optical gain, light-emitting diodes, and field-field-transistors due to the tunability of optical and electrical properties brought by the versatility of organic chemistry synthesis. Most of the works focus on achieving a higher power conversion efficiency and/or light-emitting properties through a variety of chemical synthesis, novel growth conditions, and fabrication methods [1, 2]. The solution-processed three-dimensional (3D) organic-inorganic halide perovskites have shown great promise as solar cells [1] due to high charge carrier mobility, long exciton diffusion length, and low concentration of traps, but their poor stability over longer periods of time due to heat, moisture, light, etc. has stopped them from becoming widely commercialized. However, two-dimensional perovskites (2D) have emerged as a replacement for the 3D perovskites, offering superior properties such as longer lifetime, more stability, higher bandgap, and versatility of organic chemistry synthesis [3-7]. However, the studies on the electronic structure and properties of 2D perovskite materials are very limited. Therefore, the investigation of optoelectronic properties in relation to chemical synthesis and morphological changes is critically important. In this work, we successfully synthesized deuterated methylammonium iodide (CH_3ND_3I) and prepared deuterated 2D perovskite $(CH_3ND_3)_2Pb(SCN)_2I_2$ thin films and studied the impact of deuteration on morphological changes. X-ray diffraction (XRD) measurements were carried out for the structural characterization and scanning electron microscopy (SEM) was used for morphological characterization.

Methylammonium iodide (CH_3NH_3I), Lead Thiocyanate ($Pb(SCN)_2$), Deuterium Oxide (D_2O), Chlorobenzene (CB), and N-N-dimethylformamide (DMF) were purchased from Sigma-Aldrich. The deuterated sample of CH_3NH_3I was prepared by dissolving CH_3NH_3I (0.1589) grams into 20 ml of D_2O in Round Bottom Flask and then dried at 60 °C using Rotary Evaporator. The weak acid nature of the ammonium group of CH_3NH_3I allows the exchange of protons with deuterons from D2O. The process was then repeated 3 times to ensure complete substitution of hydrogen into deuterium. Once CH_3ND_3I was produced, the precursor solution for $(CH_3ND_3)Pb(SCN)_2I_2$ was prepared by dissolving 0.220 g of CH_3ND_3I and 0.224 g of $Pb(SCN)_2$ in 1 mL of DMF. The glass substrates were cleaned ultrasonically using deionized water and soap, isopropanol, acetone, and deionized water for 15 minutes per cycle before the film was prepared. The thin films were prepared by spin-casting 100-150 mL of solution on glass substrate for 40 seconds, dropping 800 mL of chlorobenzene after 7 seconds. The XRD measurements were conducted on thin films using a Rigaku Miniflex 600 XRD machine. For SEM, thin films were sputter-coated with gold and imaged in a JEOL JSM-6510LV SEM.

Figure 1 shows the XRD patterns of $(CH_3NH_3)Pb(SCN)_2I_2$ and $((CH_3ND_3)Pb(SCN)_2I_2$ perovskite thin films in the scan range 5-60°. The typical peaks of $CH_3(NH_3)_2Pb(SCN)_2I_2$ sample are within

this range and are consistent with the previously published results [8]. Evenly spaced characteristic peaks at 9.4°, 19.0°, and 28.8° are observed in $(CH_3NH_3)Pb(SCN)_2I_2$ which are attributed to the $(CH_3NH_3)Pb(SCN)_2I_2$ phase. The same peak positions verify that deuteration didn't alter the crystal structure of $CH_3(NH_3)_2Pb(SCN)I_2$. **Figure 2** shows the SEM images of $CH_3(NH_3)_2Pb(SCN)_2I_2$ and $(CH_3ND_3)_2Pb(SCN)_2I_2$ thin films prepared under similar conditions. We observed morphological changes in $CH_3(NH_3)_2Pb(SCN)I_2$ upon deuteration. There is formation of two-dimensional crystallites in both films. However, the grain size was different in these films (**Figure 2**). This suggests that morphological changes caused by the deuteration can result in the contrast in the device performance of optoelectronic devices based on 2D perovskites.

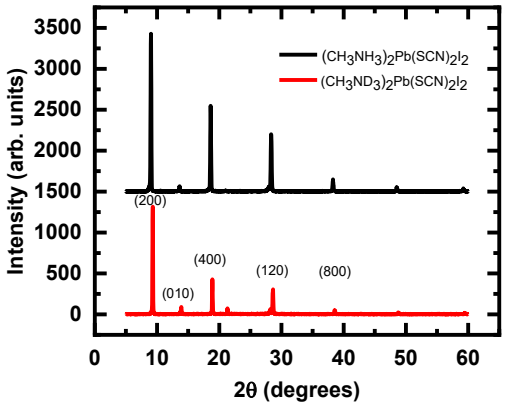

Figure 1. XRD patterns of $(CH_3NH_3)_2Pb(SCN)_2I_2$ and $(CH_3ND_3)_2Pb(SCN)_2I_2$

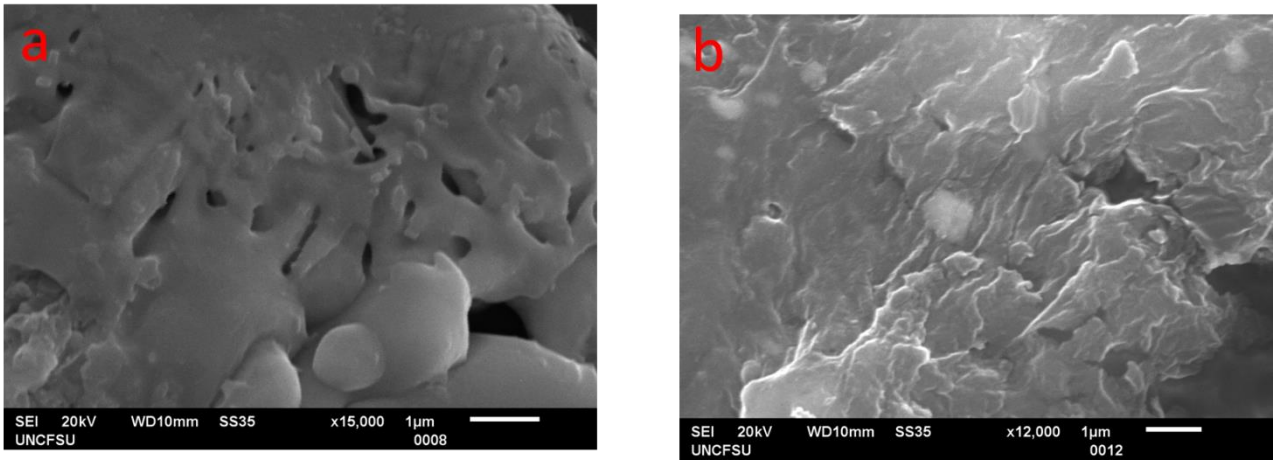

Figure 2. SEM images of $(CH_3NH_3)_2Pb(SCN)_2I_2$ (a) and $(CH_3ND_3)_2Pb(SCN)_2I_2$ (b)

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