Experimental report

12/01/2018

Proposal: **1-04-117** **Council:** 10/2016

Title: Stabilizing hybrid organometal halide perovskite films against moisture by using a sandwich structure approach

Research area: Materials

This proposal is a continuation of 1-04-99

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Samples: (CH3NH3+)[PbI3]

Instrument	Requested days	Allocated days	From	To
D22	2	2	09/02/2017	11/02/2017

Abstract:

Using hybrid organometal halide perovskites as sensitizer in hybrid solar cells has attracted very high attention in the last two years due to the very high efficiencies which have been reached within this time. One of the remaining major challenges is the moisture sensitivity of the used perovskites, which seriously limits application in solar cells since under the influence of moisture the solar cells rapidly degrade (within minutes). Based on previous experiments at D22 (see report experiment 1-04-99) giving insights into moisture ingression, we realized a stabilization by using a sandwich structure that has a thin layer of 2D perovskite on top of a thick film of 3D perovskite. In the present investigation we want to gain deeper insights into this very promising stabilization mechanism. We want to follow the ingression of moisture into such sandwich architectures as function of the thickness of the protecting 2D perovskite top layer with in-situ grazing incidence small angle neutron scattering (GISANS). The thickness of the layer of 2D perovskite top layer will be varied.

Stabilizing hybrid organometal halide perovskite films against moisture by using a sandwich structure approach
(Exp-No. 1-04-117)

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Hybrid lead halide perovskites have revolutionized the field of thin film optoelectronics by combining easy fabrication from solution chemistry with high performance when used as active material in photovoltaics. State-of-the-art devices achieve power conversion efficiencies over 22 % with an optimized quaternary cation composition (denoted FAMARC).[1] A remaining issue is the fast degradation of hybrid perovskites in ambient moisture. Exchanging the organic cations with longer organic molecules leads to the formation of 2D structures that possess higher moisture stability, but with cutbacks in device efficiency.[2] A promising approach by Hu et al. combines high photovoltaic performance of 3D perovskites with the good moisture resistance of 2D perovskites by a sandwich approach that has a thin layer of 2D perovskite on top of a thick film of 3D perovskite.[3]

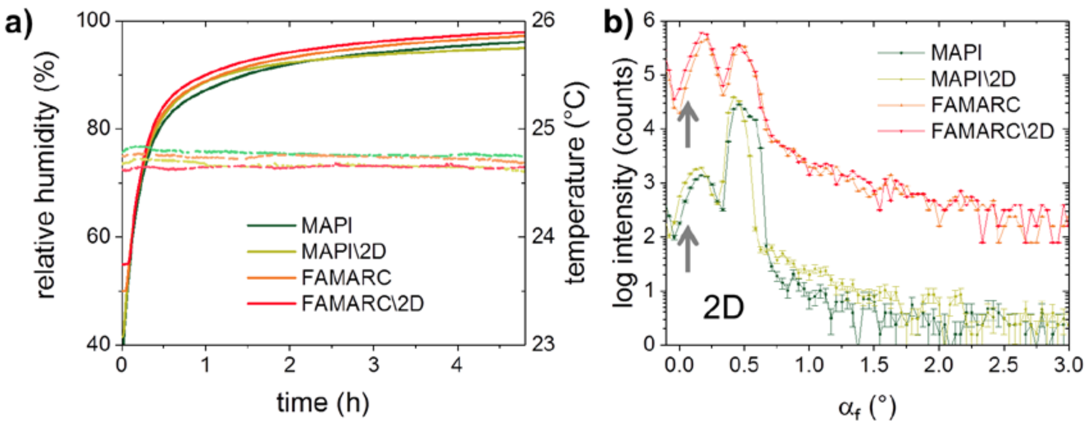

FIGURE 1: *(a) Humidity trends present in the experimental chamber during the in-situ GISANS degradation studies of perovskite thin films show that high humidity of over 90 %RH is established after 1 h, while temperature is kept constant. (b) Vertical line cuts of 2D GISANS data for all probed samples. The 2D top-layer is embodied in the bulge left of the Yoneda peaks as compared to the plain films. Data is shifted for clarity.*

Here, the moisture stability of a conventional MAPI film is tested in a plain film and one with a 2D protection layer (MAPI\2D). Following the success of our previous experiment at D22, we use in-situ grazing incidence small angle neutron scattering (GISANS) in combination with deuterated water (D_2O) to follow the ingression of moisture into perovskite films.[4] A custom aluminum chamber including a basin for the water is used to establish a stable and controlled high relative humidity of > 90%RH. As the FAMARC composition was shown to be especially sensitive to humidity, we also compare a plain FAMARC film with one with the same 2D perovskite formulation as protection layer.[5]

Figure 1 shows the humidity trends and vertical (with respect to sample surface) line cuts of the 2D GISANS data for the four studied films. A bulge at lower angles next to the Yoneda peak indicates the presence of the 2D layer in MAPI\2D (light green) and FAMARC\2D (red) samples.

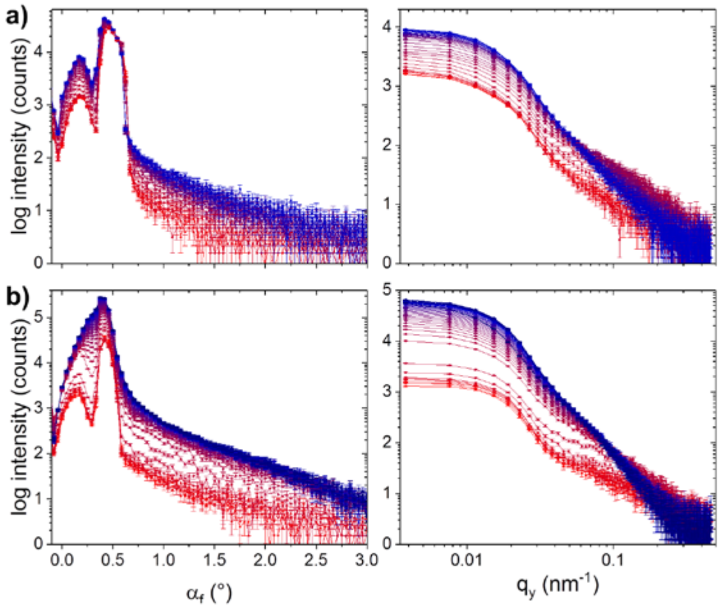

FIGURE 2: *In-situ data collected for (a) MAPI and (b) MAPI\2D. In the vertical line cuts on the left the position of the Yoneda peak only changes for the latter. However, morphological changes are obvious from the horizontal line cuts (right) for both samples.*

Surprisingly, all films showed degradation, the severest (as expected) in the FAMARC compounds. A comparison of the plain MAPI film with the MAPI\2D seems to imply a stronger degradation in the film with protection layer (cf. Figure 2). However, model fits of the data reveal that only the smallest structures in the MAPI\2D deteriorate, and as they are not present in the plain MAPI film, it can be interpreted as the 2D layer being "sacrificed" while the 3D perovskite film underneath is preserved. This important finding helps to understand the protection mechanism of the capping layer.

ILL personnel (Lionel Porcar) will be coauthoring publication(s) resulting from the experiments.

[1] M. Saliba, T. Matsui, K. Domanski, J.-Y. Seo, A. Ummadisingu, S. M. Zakeeruddin, J.-P. Correa-Baena, W. R. Tress, A. Abate, A. Hagfeldt, M. Grätzel, *Science* **2016**, *354*, 206.

[2] Y. Chen, Y. Sun, J. Peng, J. Tang, K. Zheng, Z. Liang, *Adv. Mater.* **2018**, *30*, 1703487.

[3] Y. Hu, J. Schlipf, M. Wussler, M. L. Petrus, W. Jaegermann, T. Bein, P. Muller-Buschbaum, P. Docampo, *ACS Nano* **2016**, *10*, 5999.

[4] P. Müller-Buschbaum, L. Bießmann, E. Metwalli, L. Oesinghaus, L. Porcar, J. Schlipf **2016**, *Institut Laue-Langevin (ILL)*, DOI: 10.5291/ILL-DATA.1-04-99.

[5] Y. Hu, M. F. Aygüler, M. L. Petrus, T. Bein, P. Docampo, *ACS Energy Lett.* **2017**, *2*, 2212-2218.