

Characteristics of thermal imprint with perovskite layers

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Perovskites have emerged as materials with prospects for optoelectronic devices prepared from simple solution-based processes. However, the device quality highly depends on the crystallinity and therefore on the preparation. A popular chemical route to improve the crystallinity is solvent engineering¹. A competitive mechanical route is based on thermal imprint, the planar hot pressing (PHP) with an un-patterned stamp. Current investigations have shown that PHP affects the phase change behavior², the optical properties^{3,4} and the texture of the layer^{3,5}.

This study deals with the quality improvement of perovskite layers obtained by PHP, here MAPbBr₃. Samples with distinctly different textures are subjected to an imprint process at 150°C, a temperature which appears convenient from previous experiments⁴. The different textures are obtained by varying the temperature during the soft bake of the spin-coated layer (Fig. 1). Inspection of these pristine layers (Fig. 2) reveals that the crystallites exhibit an orientation of $90^\circ \pm 20^\circ$ with respect to the substrate. At times cracks appear at low soft bake temperatures; these cracks are healed during the PHP process (Fig. 3). In consequence, PHP is capable of ‘equalizing’ the perovskite layer not only in terms of thickness and crystallinity but also in terms of defectivity (cracks, holes). In this respect we explore the impact of the pressure, temperature and time of the PHP process. The layer quality is characterized by SEM, XRD, AFM and optical measurements.

To investigate the crystallization of the perovskite in detail the imprint machine is loaded after heat-up only (hot loading). In that way very short and well-defined PHP times can be achieved. Fig. 4 exemplifies the temporal evolution of the perovskite texture/morphology, starting from a pristine layer and ending with a layer after 3 min PHP. Already after 30 s micrometer-sized crystals are formed, alternating with holes surrounded by less dense regions; these become integrated into the larger crystals with time. We will present details of this study.

All experiments are designed in a way that the samples are only imprinted in parts. Comparing the imprinted and non-imprinted regions hence allows direct conclusions on the impact of pressure during PHP.

¹ M. Xiao et al, *Ang. Chem.* **126**, 10056 (2014); T. Ma et al, *J. Phys. Chem. Lett.* **8**, 720 (2017).

² L. Dimesso et al, *J. Mater. Sci.* **54**, 2001 (2019).

³ J. Xiao et al, *J. Mater. Chem.* **A 3**, 5289 (2015).

⁴ N. Pourdavoud et al, *Adv. Mater. Technol.* **3**, 1700253 (2018); *Adv. Mater.* **29**, 1605003 (2017).

⁵ A. Mayer et al, *J. Vac. Sci. Technol.* **B 35**, 06G803 (2017).

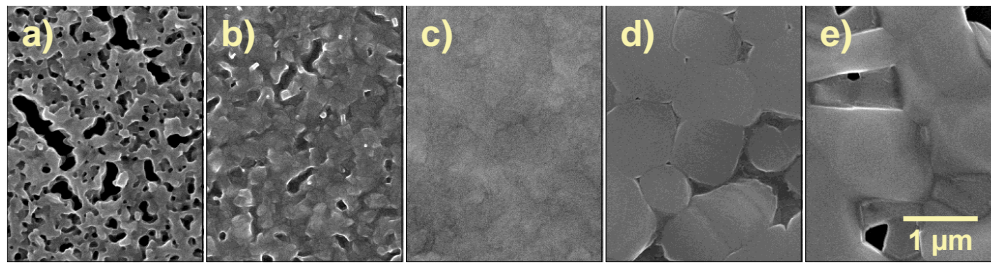


Figure 1: Pristine layers (MAPbBr_3) of different texture – Soft bake conditions: 2 min at a) 75°C, b) 100°C, c) 125°C, d) 150°C and e) 200°C; solvent DMF.

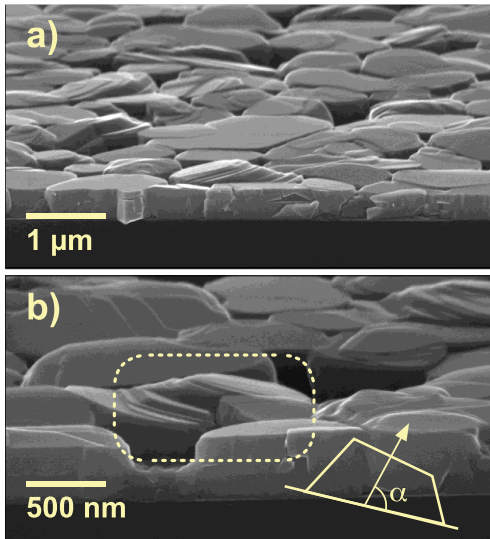


Figure 2: Crystallite orientation – a) cross-section of pristine layer, b) detail and evaluation.

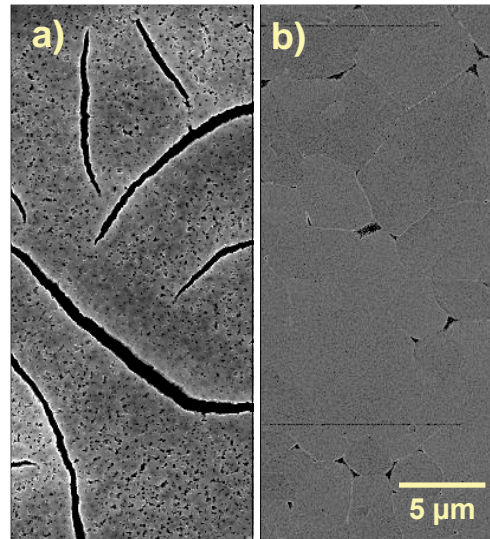


Figure 3: Healing of cracks by PHP – a) pristine layer with cracks, b) crack-free layer after PHP.

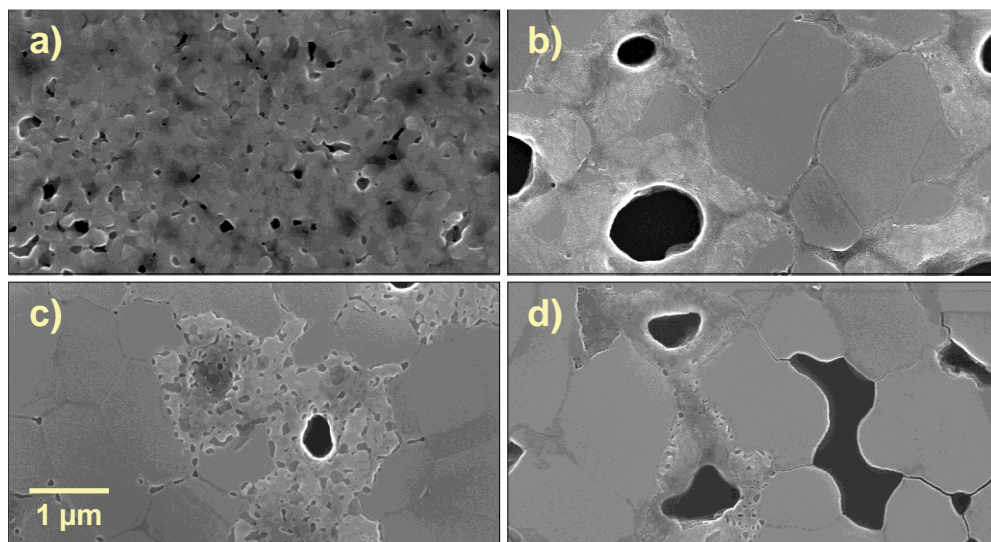


Figure 4: Crystallization during planar hot pressing PHP (150°C, hot loading) – a) pristine layer (75°C, 2min) b) after 30s, c) after 1 min and d) after 3 min PHP.