

In-situ and in-operando investigations on slot-die printed perovskite thin-films for photovoltaic application

Thin-film organic-inorganic metal halide perovskite based solar cells (PSCs) are researched intensively for their potential as cheap, ultra-thin and flexible next-generation solar cells. Reaching module efficiencies of around 17% and 15% on ultrathin flexible substrates, commercialization is promising and the efficiency can compete with silicon-based solar cells, which are currently dominating the world-wide photovoltaic (PV) market.^[1]

However, there is much potential to further improve perovskite based solar cells. In our group we focus on elucidating [degradation mechanisms](#), tuning and [finding new absorber materials](#) and shedding light on the [dynamic processes in the absorber](#).

Slot-die printing of perovskite absorber layers

Additionally, the development of industrial compatible deposition methods with maintained efficiencies requires further research and development.^[2] In principle, industrial requirements for high throughput and cheap solar cell production can be met by roll-to-roll slot-die coating.^[3] An introduction to slot-die coating is e.g. available from Ossila.^[4]

In our group, we work with our self-built slot-die coater in order to investigate and optimize printed absorber layers and fabricate printed PSCs.

Probing crystal structure in thin-films with X-rays (GIWAXS)

Thin-film quality is paramount for high efficiency PSCs. Crystal structure, morphology and texture of the absorber thin-film play an important role.^[5] Especially investigating phase formation and phase transitions over time (in-situ) can reveal the physical and chemical processes and mechanisms during film formation or degradation.

To access in-situ crystallographic information about thin-films we apply grazing-incidence wide-angle X-ray scattering (GIWAXS). This technique is well suited to probe the influence of temperature, ambient moisture, printing parameters and precursor composition on crystallization kinetics and crystal film quality by probing stochastically relevant large sample areas.^[6-8] It accesses typical length scales of the sample in the atomic distance regime, thus revealing changes in the crystal structure. More information about the technique can be found [here](#).

In-situ GIWAXS on slot-die printed perovskite films

With high brilliance synchrotron radiation sources it is possible to achieve a time resolution of less than 1 s. Thus, transient evolution of e.g. phase transitions and texture can be observed in real time. Corrected 2D GIWAXS images of spin casted and printed perovskite films are exemplary shown in fig. 1 before and after the annealing process. Especially in the low q-regime pronounced crystallographic changes are apparent, stronger evidence is visible in the 2D-plot of azimuthally integrated images. The printing was carried out at ambient conditions with a self-built slot-die coater which was equipped with a meniscus guiding blade and temperature controlled sample stage.

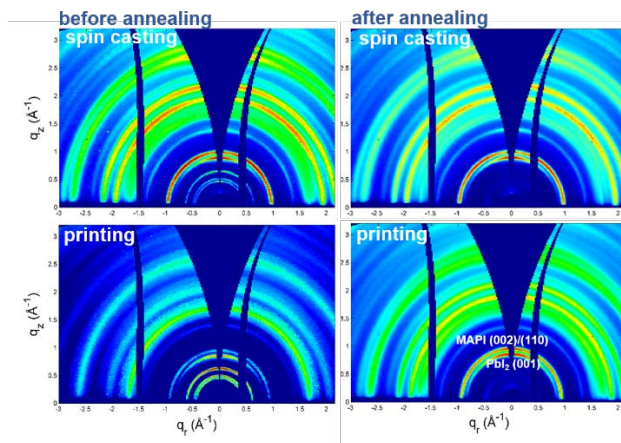


Fig. 1: 2D-GIWAXS images before and after the annealing process for spin casting and printing deposition.

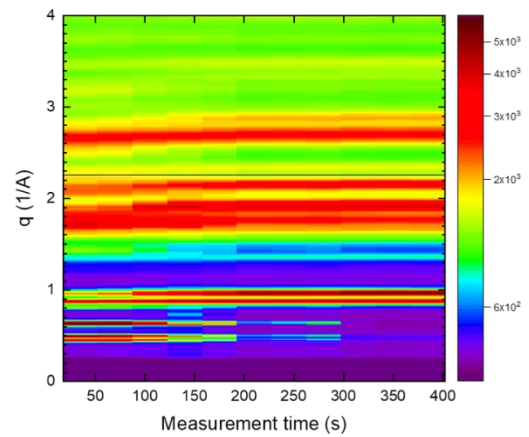


Fig. 2: 2D-plot of azimuthally integrated GIWAXS images show the time dependent crystal structure evolution during the annealing process of the slot-die printed sample.

In-operando GIWAXS and Current-Voltage measurements

Various perovskite compositions and deposition techniques are currently investigated. Many of them are adaptable to slot-die coating. Interesting insights into the rich material class of perovskites can be achieved by comparing slot-die coated samples with e.g. spin-coated samples.^[9] Understanding how to tune texture and morphology by e.g. optimizing underlying layers and deposition techniques, can significantly decrease losses and boost the power conversion efficiency (PCE). In-operando current-voltage (IV) and GIWAXS experiments have been performed at high brilliance synchrotron radiation facilities to find correlations between crystal structure, texture, morphology and IV cell parameters.

Highlighted Works

- Toward Tailored Film Morphologies: The Origin of Crystal Orientation in Hybrid Perovskite Thin Films^[9]
- Shedding Light on the Moisture Stability of 3D/2D Hybrid Perovskite Heterojunction Thin Films^[10]
- Single-crystal-like optoelectronic-properties of MAPbI₃ perovskite polycrystalline thin films^[11]
- Structure of Organometal Halide Perovskite Films as Determined with Grazing-Incidence X-Ray Scattering Methods^[7]
- Capturing the Sun: A Review of the Challenges and Perspectives of Perovskite Solar Cells^[2]
- Nanostructuring Mixed-Dimensional Perovskites: A Route Toward Tunable, Efficient Photovoltaics^[12]

References

- [1] M. A. Green, Y. Hishikawa, E. D. Dunlop, et al., *Progress in Photovoltaics: Research and Applications* **2019**, 27, 3-12.
- [2] M. L. Petrus, J. Schlipf, C. Li, et al., *Advanced Energy Materials* **2017**, 7.

- [3] Y. Rong, Y. Ming, W. Ji, et al., *J Phys Chem Lett* **2018**, 9, 2707-2713.
- [4] Ossila, *Slot-Die Coating: Theory, Design, & Applications*, <https://www.ossila.com/pages/slot-die-coating-theory>, **2020**.
- [5] Y.-S. Jung, K. Hwang, Y.-J. Heo, et al., *Advanced Optical Materials* **2018**, 6.
- [6] G. E. Eperon, V. M. Burlakov, P. Docampo, et al., *Advanced Functional Materials* **2014**, 24, 151-157.
- [7] J. Schlipf, P. Müller-Buschbaum, *Advanced Energy Materials* **2017**, 7.
- [8] J. Schlipf, L. Biessmann, L. Oesinghaus, et al., *J Phys Chem Lett* **2018**, 9, 2015-2021.
- [9] L. Oesinghaus, J. Schlipf, N. Giesbrecht, et al., *Advanced Materials Interfaces* **2016**, 3.
- [10] J. Schlipf, Y. Hu, S. Pratap, et al., *ACS Applied Energy Materials* **2019**, 2, 1011-1018.
- [11] N. Giesbrecht, J. Schlipf, I. Grill, et al., *Journal of Materials Chemistry A* **2018**, 6, 4822-4828.
- [12] T. M. Koh, V. Shanmugam, J. Schlipf, et al., *Adv Mater* **2016**, 28, 3653-3661.