Sequential Evaporation of Inverted $FAPbI_3$ Perovskite Solar Cells – Impact of Substrate on Crystallization and Film Formation

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Abstract

Recent advances in sequential evaporation of perovskite solar cells (PSCs) have culminated in a rapid increase in reported power conversion efficiencies (PCEs), now on par with the best solution-processed counterparts. This development triggered vast interest of industry and academics. To date, however, very few studies addressed sequentially evaporated PSCs in the *p-i-n* architecture and an in-depth process understanding is lacking. Here, we investigate the impact of the hole transport layer (HTL) on the formation of formamidinium lead triiodide (FAPI) perovskite thin films fabricated via an evaporated two-step process. We find that the crystal orientation of lead iodide (PbI_2) changes significantly for different HTLs, thereby impacting the subsequent conversion and crystallization process. Adjusting the amount of deposited FAI reveals an unexpected correlation of the PbI_2-to-perovskite X-ray diffraction peak intensity ratio to final PSC performance that depends on the employed HTL. Our approach enables PCEs of more than 17%, the highest reported for fully vacuum-processed pure FAPI PSCs in the *p-i-n* architecture.

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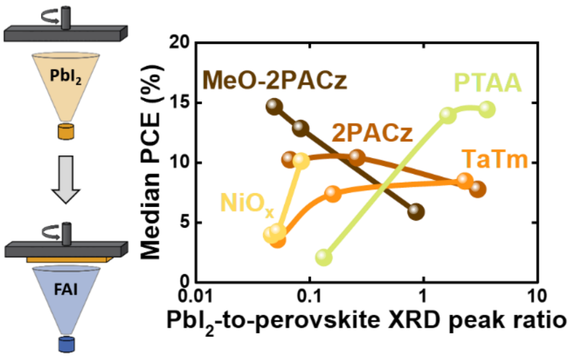

Organic-inorganic metal halide perovskite solar cells (PSCs) are one of the most promising options for next-generation photovoltaics, given their tunable band gap,[1,2] long charge carrier diffusion lengths,[3] defect tolerance, and high absorption coefficients.[4] The rapid increase in reported power conversion efficiencies (PCEs) over the past decade, with certified PCEs approaching 27%, made them serious competitors to established thin-film technologies such as copper indium gallium-selenide (CIGS) and cadmium telluride (CdTe).[5,6] Most research on PSCs has focused on solution-based fabrication methods of the perovskite absorber layer (*e.g.*, spin coating, inkjet printing, blade coating, and slot-die coating).[7] These processes allow for rapid and cost-effective process optimization and fast perovskite layer deposition. In contrast, less than 1% of all existing publications report on PSCs fabricated using vacuum-based thermal evaporation (TE) deposition processes even though they dominate today's established thin-film manufacturing.[8–10] Furthermore, TE offers conceptual advantages such as homogeneous deposition over larger areas and conformal coverage of textured surfaces for industrially relevant applications.[11–13] Most reports on TE employ a co-evaporation approach, in which all perovskite precursor materials are deposited simultaneously in a single deposition step. The first reports of co-evaporated PSCs in 2013 already achieved PCEs of up to 15.4%, employing methylammonium iodide (MAI) and lead chloride ($PbCl_2$) as precursors.[14] Since then, more complex compositions (*e.g.*, double-cation, triple-cation and mixed halides)[15–18] as well as more advanced processes (*e.g.*, faster deposition, multi-halide molten salts)[19–22] have increased our understanding of TE processes significantly. The current record PCE of co-evaporated PSCs reported by Leyden *et al.* in 2024 is 21%[23] – significantly below the values achieved for solution processing.[8] One reason for the limited PCE is that co-evaporated perovskite thin films exhibit a pronounced substrate dependency, complicating process optimization. There are several studies that address the complex impact of the substrates on perovskite formation during co-evaporation.[24–29] In our previous study, Abzieher *et al.* showed the effect of different HTLs on co-evaporated methylammonium lead triiodide (MAPI) perovskite thin film formation and correlated the observations to different surface polarities of the substrates.[27] Furthermore, interfacial interactions such as the formation of hydrogen bonds between phosphonic acid functional groups in case of self-assembled monolayer (SAM)-based HTLs and interfacial iodide anions impact reaction kinetics for co-evaporated formamidinium (FA)-based absorbers, resulting in increased organic incorporation rate and preferential α-$FAPbI_3$ growth.[24,25,29]

An alternative vacuum-based TE method is the sequential layer deposition process, where the precursor materials are deposited in two or more subsequent deposition steps. The first study by Chen *et al.* investigating this process employed $PbCl_2$ and MAI as precursor materials and achieved a decent PCE of 15.4% in 2014.[30] The following years exhibited increases in reported PCEs reaching up to 21.3% in 2021 for a $FA_xCs_{1-x}PbI_3$ absorber.[31–33] More recent studies by Yi *et al.* proved the great potential of the sequential layer deposition approach by reaching PCEs of 24.4% in 2023 and over 26% in 2024, closing the gap to solution-processed PSCs (see Figure 1b).[20,34,35] Various approaches have been reported, such as single-layer and alternating multilayer depositions, as well as the use of different material compositions of the perovskite layer, demonstrating the versatility and applicability of this fabrication approach.[31–33,36–38] However, most studies on the vacuum-based sequential layer deposition process focus on the *n-i-p* architecture,[20,31,32,34,36,38–43] while reports on the *p-i-n* architecture are heavily underrepresented with a highest reported PCE of 19.4% for pure $MAPbI_3$ PSCs (see Figure 1c and Table S1).[30,33,37,44,45] The *p-i-n* architecture displays several key advantages in the context of future applications, *e.g.*, facile integration into monolithic tandem PV devices and high operational stability, making further research and development crucial.[46–49] Decoupling the simultaneous co-evaporation of precursor materials into two or more subsequent deposition steps promises several advantages: (i) the ability to reduce cross-contamination and thus improve reproducibility since different evaporation chambers are used for each material, and (ii) a straight-forward integration to industrial scale in-line processing using several linear evaporators with greater potential for high-throughput as compared to co-evaporation.[50] While the impact of the substrate on the crystallization of co-evaporated perovskites is well studied,[24,27,28] the interplay between the substrate and the quality of sequentially evaporated perovskite thin films has not been investigated yet.

In response, this work investigates the effect of different HTLs on the formation of formamidinium lead triiodide (FAPI) perovskite thin films fabricated via an evaporated two-step process. We observe significant differences in the morphology and crystallinity of the films, impacting the performance of resultant FAPI PSCs in the *p-i-n* architecture. To identify the reason for this, we analyze the firstly deposited PbI_2 layers using scanning electron microscopy (SEM), atomic force microscopy (AFM), and grazing-incidence wide-angle X-ray scattering (GIWAXS) measurements. We find a strong impact of the HTL on the morphological properties of the PbI_2 layer and reveal a significant change in crystal orientation, which subsequently affects the conversion to the perovskite phase after deposition of FAI. To further investigate the relationship of different PbI_2 structural properties and conversion to the final perovskite phase, we adjust the FAI to PbI_2 stoichiometry and find an unexpected correlation of the X-ray diffraction (XRD) pattern and final PSC performance that serves as a guideline for PCE optimization on different HTLs. Finally, we show the possibility of manipulating the observed structural properties by adjusting process parameters (evaporation rate and substrate temperature) during the deposition of the PbI_2 layer. We achieve PCEs of up to 17.2% (stabilized at 16.0%), the highest reported number for a fully vacuum-processed pure FAPI perovskite composition in the *p-i-n* architecture. In summary, this work provides an in-depth understanding of the effect of substrate for the vacuum-based sequential evaporation process and emphasizes the difficulty to analyze and compare 2D XRD patterns of the same perovskite composition on different HTLs.

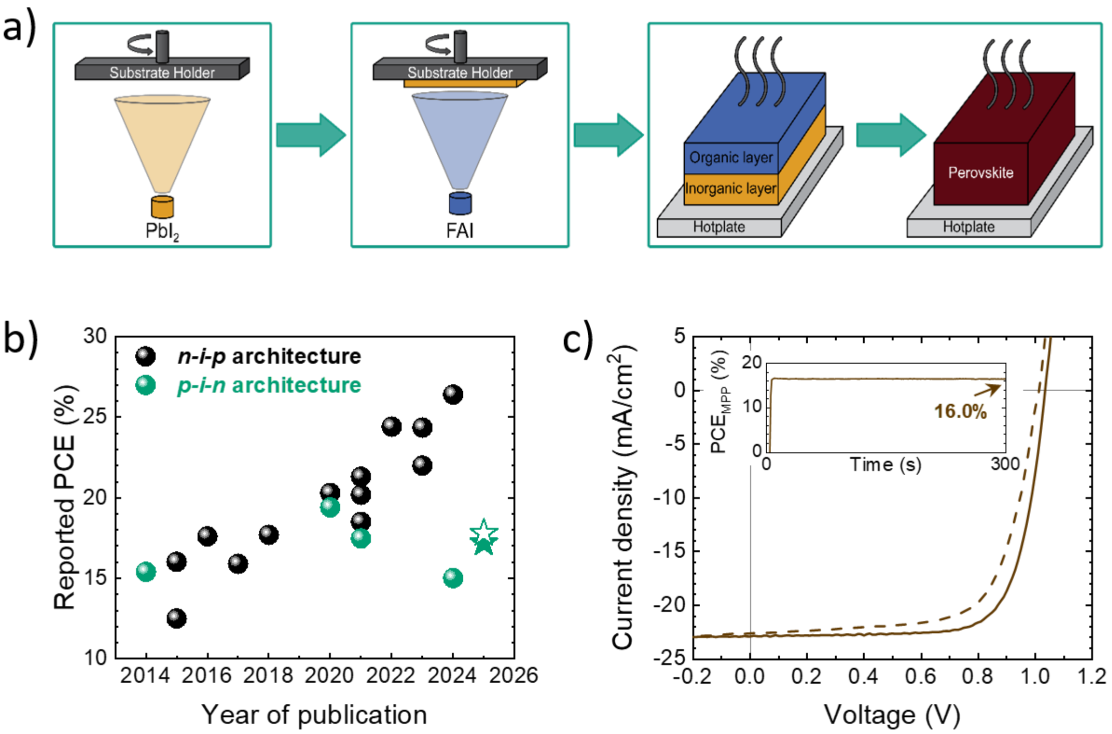

Figure 1. a) Schematic of the sequential layer deposition process. b) Published champion PCE values of sequential layer deposition processes (see more details in Table S1). The stars represent the champion PCEs for FAPI (filled) and $FA_{0.85}Cs_{0.15}PbI_3$ (empty) presented in this work. c) J-V analysis and MPP-tracking of the champion device with the layer sequence ITO/evaporated MeO-2PACz/perovskite/C_{60}/SnO_x/Ag.

Vacuum-based sequential layer deposition of perovskite thin films encompasses a wide range of deposition sequences. Existing reports vary from subsequent single-material layer depositions to alternating multilayer deposition processes, as well as processes using co-evaporated intermediate steps.[30–34,36–38,51,52] The various reported processes and resulting photovoltaic parameters are summarized in Table S1. Here, we apply an evaporated two-step process to fabricate FAPI PSCs in the *p-i-n* architecture as schematically depicted in Figure 1a. In the first evaporation step we deposit the PbI_2 layer, followed by the deposition of FAI in the second evaporation step (see Experimental Section in the SI for further details). Both deposition processes are carried out in dedicated evaporation chambers to avoid cross-contamination. Conversion to the final perovskite phase happens during a subsequent annealing step under ambient atmosphere (~30-40% relative humidity), with humidity enhancing diffusion of the organic cations.[34,43,52] In this work we used optimized annealing parameters of 170 °C and 15 min, which is in line with reports from literature working with similar processes.[20,34,45,51] The *J-V* characteristics of the best performing PSC are displayed in Figure 1c. It consists of the layer stack glass/ITO/MeO-2PACz/FAPI/C_{60}/ALD-SnO_x/Ag, using our recently developed evaporated MeO-2PACz SAM-HTL (from now on referred to as MeO), with a final perovskite film thickness of 500 nm.[53] The PCE in the reverse scan is 17.2% (J_{SC} = 22.9 mA/cm^2, V_{OC} = 1.03 V, FF = 74.2%, hysteresis Index (HI) = 0.09) with a stabilized PCE of 16.0% under maximum power point (MPP) tracking, the highest reported PCE of fully vacuum-processed FAPI PSCs in the *p-i-n* architecture. We highlight that the champion cell remains functioning without a notable decrease in performance after storage in a nitrogen-filled glovebox for 10 months, showing a PCE of 16.7% in the reverse scan, a stabilized PCE of 16.2% and a reduced HI of 0.04 (see Figure S1). Having developed this recipe, we further tested whether addition of 15% Cs to our inorganic scaffold leads to an improvement in device performance. We achieved PCEs of up to 17.8% in the reverse *J-V* scan with a stabilized PCE of 16.5% (see *J-V* analysis, EQE, MPP and XRD measurements in Figure S2 and statistics in Figure S3). A summary of the photovoltaic parameters of our champion cells for FAPI and $FA_{0.85}Cs_{0.15}PbI_3$ PSCs is shown in Table 1. However, the focus in this work is on the pure FAPI composition.

Table 1. Photovoltaic parameters of champion cells for FAPI and $FA_{0.85}Cs_{0.15}PbI_3$ PSCs. The reverse J-V scan is shown in bold and the forward scan in italic. Stabilized PCE represents the PCE after 300 s of MPP tracking.

	PCE [%]	J_{SC} [mA/cm^2]	V_{OC} [V]	FF [%]	Stabilized PCE [%]
$FAPbI_3$	**17.2** *15.6*	**22.4** *22.2*	**1.03** *1.01*	**74.2** *69.6*	**16.0**
$FA_{0.85}Cs_{0.15}PbI_3$	**17.8** *15.7*	**22.6** *22.2*	**0.98** *0.97*	**80.5** *73.0*	**16.5**

To test whether differences in perovskite film formation on different HTLs occur, we investigated sequentially evaporated perovskite thin films (after annealing) by analyzing SEM and XRD measurements. The five chosen HTLs (MeO, 2PACz, TaTm, NiO_x, and PTAA) are widely used in literature and exhibit notable differences in their chemical properties. We observe changes in the average grain size of the perovskite thin film as depicted in representative SEM images in Figure 2a. Specifically, films on MeO and 2PACz exhibit more disoriented and on average smaller grains, while the average grain size is larger on TaTm, NiO_x and PTAA (see Figure S6 for detailed analysis), which can also be seen in the corresponding cross-sectional SEM images (Figure S7). This indicates that the underlying substrate directly impacts the morphology of sequentially evaporated perovskite thin films. XRD analysis of the films further supports this interpretation. Perovskite layers on all HTLs show a different ratio of the (100)-PbI_2 to (001)-perovskite peak intensities (from now on referred to as PbI_2-to-PVK peak ratio), indicating a change in the crystallization and conversion process to the perovskite phase (Figure 2b). In addition, the performance of completed PSCs differs significantly, with no correlation to the observed differences in XRD patterns (Figure 2c, Figure S4 and Figure S5). In literature, often a beneficial effect of small or even significant amounts of residual PbI_2 in the perovskite film is observed, independent of the employed HTL/ETL.[54,55] In our case, PCEs of PSCs on

MeO reach median values of around 15% with a negligible PbI_2-to-PVK peak ratio, similar to those obtained in case of PTAA as HTL which, however, exhibits the highest PbI_2-to-PVK peak ratio from XRD. This reveals that the comparison of XRD patterns of perovskite thin films on different HTLs is not a reliable indicator for the performance of complete PSCs. We note that the different HTLs themselves can have an impact on device performance due to variations in non-radiative interfacial recombination and selective hole transport.[56,57] In addition, the thermal stability of HTLs is a crucial factor for long-term stability of *p-i-n* PSCs.[58–60] Yet, the fact that all employed HTLs yield a stable PCE during MPP tracking for 5 minutes indicates that no thermal degradation occurs under our annealing conditions (see Figure S5). Therefore, it is reasonable to assume that the observed differences mainly depend on a substrate-dependent perovskite film formation and its impact on resulting film quality, which is the main subject of investigation in this study. The key difference of the two-step evaporation process as compared to its co-evaporation counterpart is the formation and crystallization process of the perovskite phase. In case of co-evaporation, all precursor materials make contact with the substrate simultaneously, leading to immediate formation of perovskite nano crystallites at the interface with the substrate during deposition.[27,61] As discussed above, co-evaporation processes are therefore known to be strongly affected by the underlying substrate, resulting in the need for an optimization of process parameters to each substrate to form perovskite films of comparable quality.[27] In contrast, perovskite film formation and crystallization in case of the evaporated two-step process starts at the interface between the subsequently evaporated inorganic and organic materials, and full crystallization to the perovskite phase is only achieved during the annealing step in which the inorganic and organic parts are intermixed by diffusion. Thereby, a much smaller dependence of perovskite crystal growth on the substrate is expected.

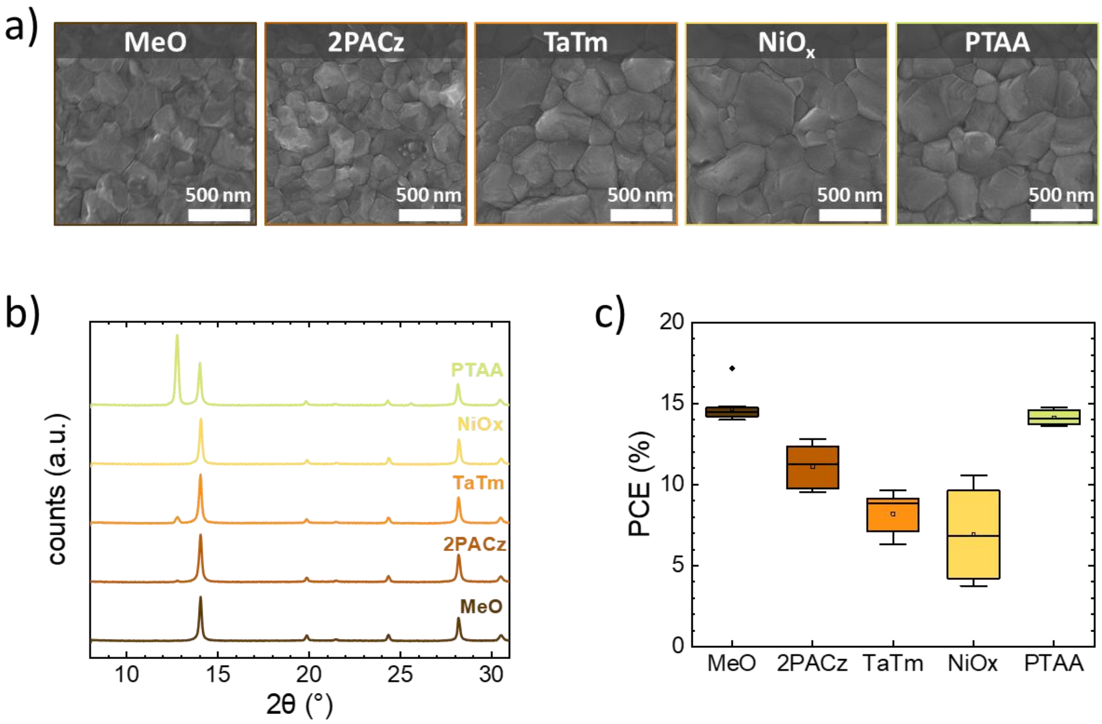

Figure 2. a) SEM top view images of 500 nm thick perovskite thin films on different HTLs. b) XRD patterns of corresponding perovskite thin films on different HTLs. c) PCE statistics of final PSCs.

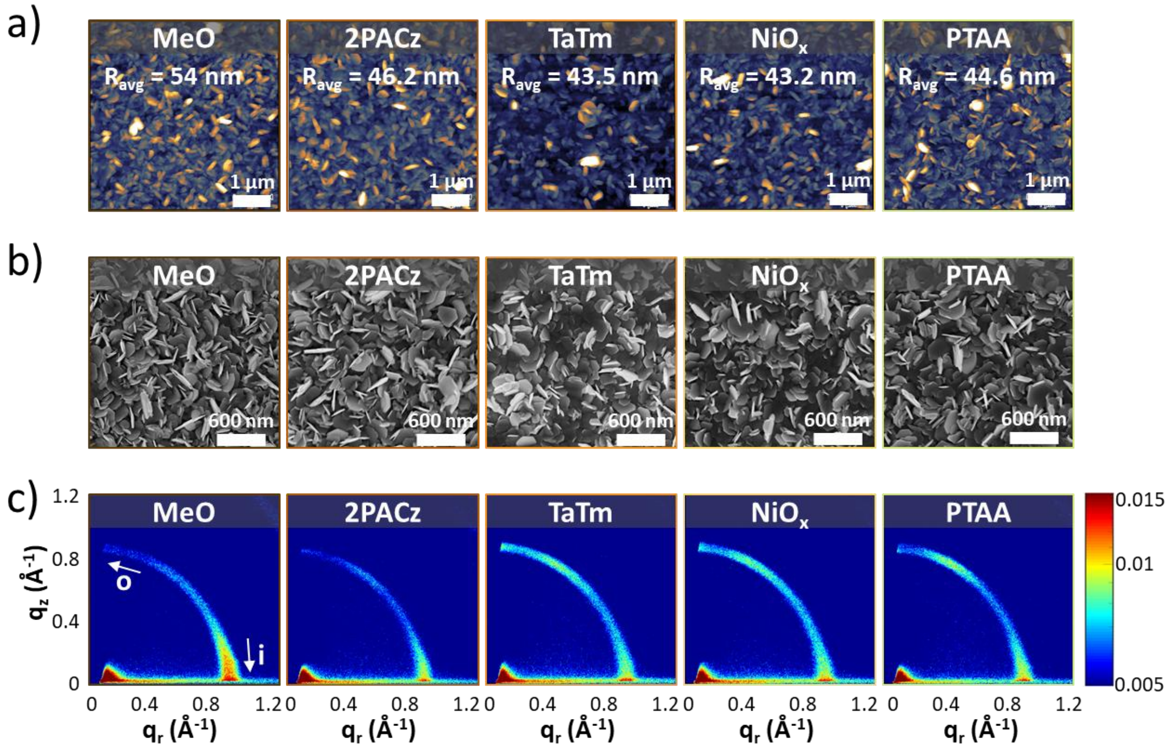

Figure 3. a) AFM analysis of 300 nm thick evaporated PbI_2 layers on different HTLs with average surface roughness values. b) SEM top view images of corresponding PbI_2 layers on different HTLs. c) GIWAXS analysis of corresponding PbI_2 layers on different HTLs. "o" denotes out-of-plane orientation, "I" denotes in-plane orientation of the crystallites.

To understand the impact of different substrates on perovskite formation in greater detail, we next analyzed the inorganic layer morphology and microstructure. From AFM measurements of the PbI_2 thin films, we find that the surface roughness (R_{avg}) on the different HTLs varies from R_{avg} = 43.2 nm in case of NiO_x to R_{avg} = 54 nm in case of MeO (see Figure 3a). A higher R_{avg} results in a larger surface area of the PbI_2 layer that is in contact with the subsequently deposited FAI. A larger contact area could simplify interdiffusion of FAI into the inorganic scaffold and accelerate the initial conversion reaction of the two precursor materials. Furthermore, in the case of dry-wet hybrid two-step deposition processes, rougher films are often considered to be more porous which facilitates the interdiffusion of the organic cations into the inorganic scaffold.[62,63] However, the differences in R_{avg} between 2PACz, TATm, NiO_x and PTAA are comparatively small, despite strong differences in final perovskite film properties and device performance, requiring more in-depth analysis. Next, we analyzed SEM images of the evaporated PbI_2 thin films on the different substrates. We observe slight changes in the orientation of the PbI_2 platelets depending on the underlying substrate (see Figure 3b and Figure S8). On MeO and 2PACz, the PbI_2 platelets appear to be more vertically oriented, which is in line with a slightly higher R_{avg} from the AFM measurements. PbI_2 layers on TaTm, NiO_x and PTAA appear to have more horizontally orientated PbI_2 platelets. We can conclude that the morphological properties, especially the orientation of the PbI_2 platelets are affected by the underlying substrate. Yet, there seems to be no clear correlation between morphological properties and final PSC performance. To further analyze the effect of different substrates on the microstructural properties of the inorganic layer, we performed GIWAXS measurements of the evaporated PbI_2 thin films. GIWAXS can give information on different crystal growth orientations and the overall degree of crystallinity of thin films.[64] PbI_2 layers on all HTLs show an intensity maximum in an in-plane orientation (referred to as "i" in Figure 3c), indicating the (001)-plane of PbI_2 is perpendicular to the substrate surface. Whereas for PbI_2 layers on MeO and 2PACz HTLs we do not detect another intensity maximum, PbI_2 layers on TaTm, NiO_x and PTAA show a second preferred orientation in a more out-of-plane orientation (referred to as "o" in Figure 3c), meaning a more vertically (001)-oriented growth. The corresponding pole figures with integrated intensity over corresponding azimuth angle are shown in Figure 4a. It has been reported previously that the crystal orientation of the PbI_2 layer can affect the diffusion of organic cations, both for the hybrid deposition and sequential evaporation processes, and therefore impact the conversion reaction during the annealing step.[34,65–69] Yi *et al.* proved that a preferred orientation of the evaporated lead iodide-chloride layer strongly affects the conversion and quality of their sequentially evaporated perovskite films.[34] However, substrate-dependent growth and the impact on crystal orientation of evaporated PbI_2 layers has not yet been investigated. More out-of-plane orientation, meaning more vertically oriented PbI_2 crystals, might facilitate the penetration and diffusion into the inorganic layer during the deposition of the organic cation and the subsequent annealing step. In order to gain further in-depth understanding of the interplay of chemical properties and resulting PbI_2 growth mechanism much more detailed characterizations are required which is beyond the scope of this study. In summary, the choice of substrate has a significant impact on the crystal growth of the inorganic layer deposited in the first step. This is evident from morphological changes such as grain size and surface roughness as well as the crystal orientation of the PbI_2 layer.

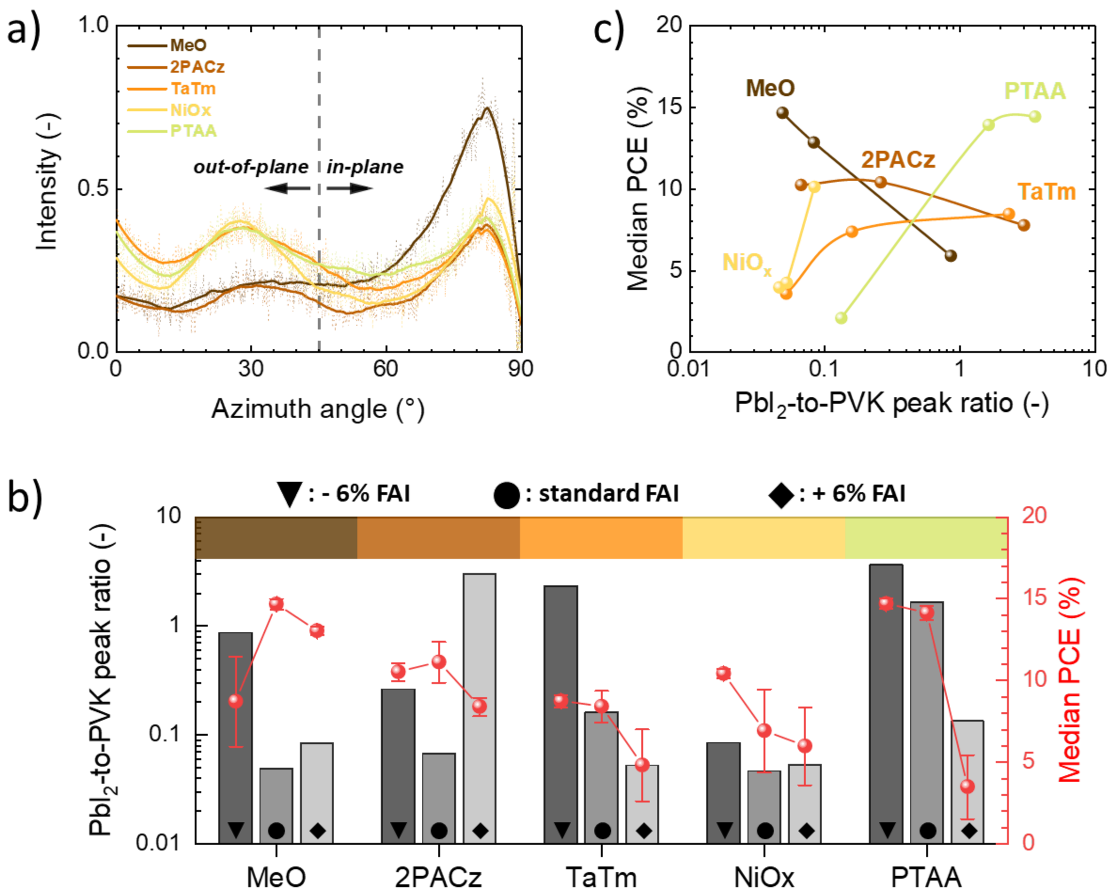

Figure 4. a) Pole figures from GIWAXS measurements of evaporated PbI_2 layers on different HTLs. b) Evolution of the PbI_2-to-PVK peak ratio for different FAI to PbI_2 stoichiometries (increasing from left to right: -6%, standard, +6%) with corresponding median PCEs. C) Guideline plot for maximum median PCE of FAPI PSCs on different HTLs with respect to the PbI_2-to-PVK peak ratio. Standard refers to deposited film thicknesses for PbI_2 and FAI of 300 nm and 280 nm respectively. The final perovskite film thickness for -6%, standard and +6% FAI is 470 nm, 500 nm and 530 nm respectively.

Next, by altering the stoichiometry of FAI to PbI_2 by adjusting the amount of deposited FAI, we reveal that the crystal orientation of the evaporated PbI_2 layer on different HTLs has a direct and unexpected impact on the conversion and crystallization process as well as the photovoltaic parameter of complete PSCs. When reducing the deposited FAI layer thickness from the standard value to -6%, we find an increased XRD PbI_2 intensity and PbI_2-to-PVK peak ratio for all studied HTLs (see Figure S9). Notably, an increase in the deposited FAI amount (+6%) shows different trends for the various HTL that seem to depend on the PbI_2 crystal orientation. Films on MeO and 2PACz with more in-plane PbI_2 orientation show an increased PbI_2-to-PVK peak ratio, in stark contrast to the expectation. PbI_2 layers with increased out-of-plane orientation show the reverse trend, leading to a further reduction of the PbI_2-to-PVK intensity ratio (see Figure 4b). Pairing these films with median PCE values of the resulting PSCs reveal a significant difference between PbI_2-to-PVK peak ratio and optimum PCE (see red data points in Figure 4b, for full statistics see Figure S10, with calculated HI for each variation see Figure S11), resulting in a direct correlation to the previously observed PbI_2 crystal orientation. PSCs on MeO and 2PACz, which show a more in-plane orientation of the firstly evaporated PbI_2 layer, display their best performing devices for a lower PbI_2-to-PVK peak ratio. In contrast, PSCs on TaTm, NiO_x and PTAA, showing more out-of-plane oriented PbI_2 crystals, yield the best results for higher relative intensities of PbI_2 in their XRD patterns (see Figure S9 and Figure S10). By plotting median PCEs over the corresponding PbI_2-to-PVK peak ratios we obtain a guideline plot showing general trends for the fabrication of sequentially evaporated PSCs and their preferred regime for optimum performance on different HTLs (see Figure 4c).

Given the importance of morphological and microstructural properties of the PbI_2 layer on the conversion to the perovskite phase, we sought to further investigate how to manipulate these properties by changing process parameters, more specifically deposition speed and substrate temperature. We chose to vary the evaporation rate of PbI_2 in the range of 0.1 Å/s to 2.0 Å/s and the temperature of the substrate (5 °C, 20 °C, 45 °C, and 70 °C) and employ MeO as SAM-HTL since it yielded the best device performance. Changing the substrate temperature for a constant PbI_2 evaporation rate of 1.0 Å/s has no major effect on the crystal orientation of the inorganic scaffold (see Figure S12a). As shown in pole figures of GIWAXS measurements, all layers exhibit a similar single orientation maximum in a more in-plane orientation (see Figure 5a). In contrast, different PbI_2 evaporation rates with a constant substrate temperature of 20 °C exhibit a clear effect on crystal orientation (see Figure S12b). While an increase in evaporation rate to 2.0 Å/s leads to a more strongly pronounced in-plane growth of the PbI_2 crystallites, slower deposition speeds display a second orientation maximum in an out-of-plane orientation (see Figure 5a). Additionally, with top view SEM images we see that both process parameters, deposition speed and substrate temperature, affect the apparent grain size of the evaporated PbI_2 layers. While slower evaporation rates result in larger PbI_2 platelets, an increase in substrate temperature leads to a decrease platelet size (see Figure S13). Both process parameters therefore do have an impact on the morphological and microstructural properties of the evaporated PbI_2 layer.

To confirm the previous findings, we used the differently processed PbI_2 layers for our evaporated two-step process to analyze the conversion to perovskite as well as the final PSC performance. After the deposition of FAI and subsequent annealing, we detect strong differences in the final XRD patterns, especially regarding the PbI_2-to-PVK peak ratio (see Figure 5b and Figure S14). The corresponding PCEs of the resulting PSCs confirm the earlier findings and are in line with the general XRD trends of our perovskite layer on MeO (compare Figure 4c). High PbI_2-to-PVK peak ratios from XRD analysis of the perovskite layer result in decreased PCEs of the corresponding PSCs, while low PbI_2-to-PVK peak ratios generally result in improved performance (see Figure 5c and Figure S15). We hereby show that it is possible to manipulate the growth and final properties of the evaporated inorganic layer simply by adapting easily accessible process parameters (deposition speed and substrate temperature) during evaporation. We observe the same trends for XRD and PSCs performance that we observed on various HTLs, underlining the validity of our guideline plot in Figure 4c.

In this work, for the first time, we reveal a substrate-dependent film formation of FAPI perovskite thin films fabricated with an evaporated two-step process. We show that the substrate affects the morphological and microstructural properties of the firstly evaporated PbI_2 layer, which impacts the conversion to perovskite after deposition of FAI and the corresponding PSC performance. Changes in crystal orientation of the PbI_2 layer as measured by GIWAXS are found to affect the PbI_2-to-PVK XRD peak intensity ratio that shows an unexpected trend, revealing that analysis of the PbI_2 peak intensity is not a reliable solar cell performance indicator. Based on these findings, we provide a guideline for XRD analysis to achieve the highest performing PSCs on different HTLs based on coupled GIWAXS and XRD analysis. Furthermore, we studied the effect of variation in process parameters on the PbI_2 layer properties and show the possibility to manipulate the PbI_2 crystal orientation by adjusting the deposition speed. Using this approach, we fabricated highly efficient fully vacuum-processed MA-free PSCs in the *p-i-n* architecture with PCEs of up to 17.2% for pure FAPI and 17.8% for $FA_{0.85}Cs_{0.15}PbI_3$. Our work further provides an in-depth understanding of the sequential layer evaporation deposition process and paves the way to highly efficient PSCs using an industrially relevant and highly promising fabrication method. Future work will focus on evaluating the advantages of this process compared to co-evaporation, particularly in context of high-throughput manufacturing using linear evaporation sources.

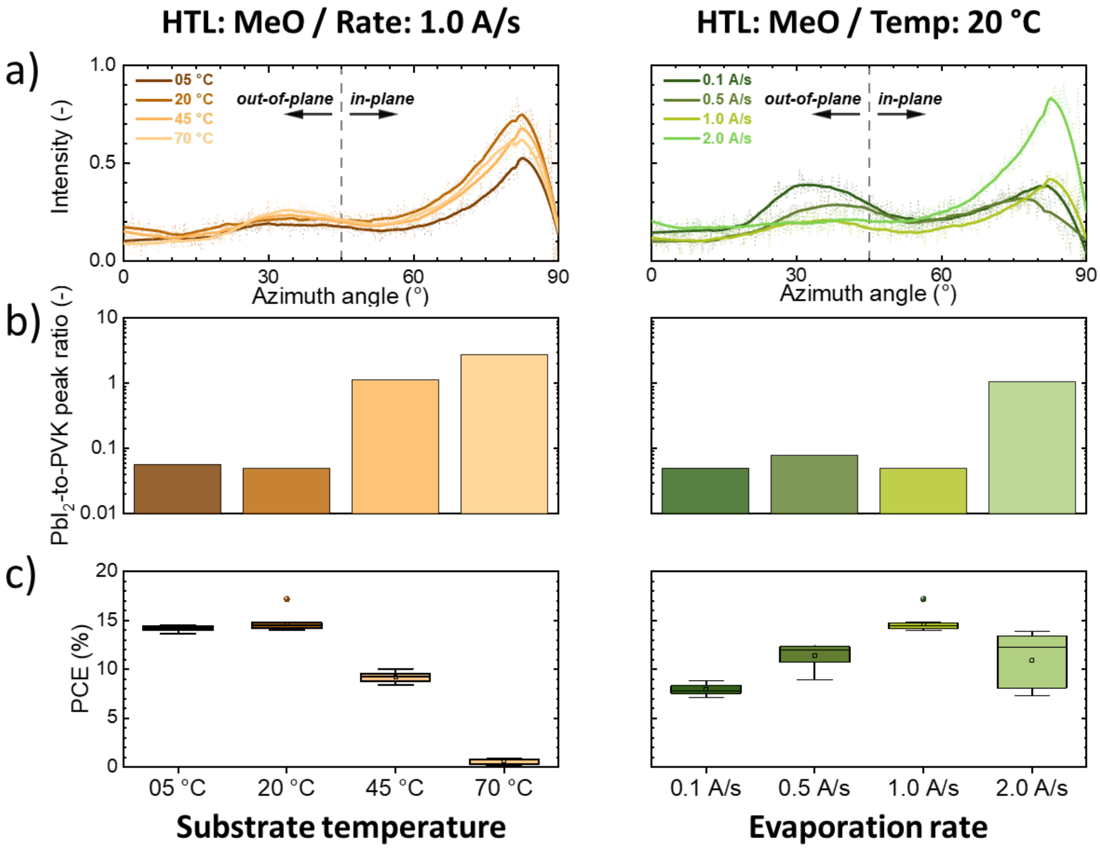

Figure 5. Analysis of varying substrate temperature (left) and PbI_2 evaporation rate (right) employing MeO as HTL. a) Pole figures from GIWAXS measurements. b) PbI_2-to-PVK peak ratios for perovskite films with standard FAI to PbI_2 stoichiometry. c) PCE statistics of respective PSCs.

Supporting Information

The Supporting Information is available free of charge:

- Detailed Experimental Section, literature analysis for sequentially evaporated PSCs, additional solar cell efficiency data, EQE, MPP, SEM, XRD, GIWAXS.

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