

Probing the in-situ dynamics of structure-property evolution in hybrid perovskite thin films spincoated from complex fluids by a custom designed, beamline compatible multimodal measurement chamber

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Hybrid perovskite materials are intriguing candidates for next-generation photovoltaic and optoelectronic applications. The formation of thin-films based on these materials is processed through chemically complex^[1], solution-based precursors. The spin-coating of complex fluids is a process far from thermodynamic equilibrium, as the dynamically transforming precursor undergoes non-trivial structural and electronic transformations throughout its conversion to the final crystalline thin-film. The structural alterations experienced by the film range from the emergence of glassy intermediate states within the drying precursor, to shear-induced morphological instabilities as a response to stresses generated in the film^[2] during drying and crystal growth. It is therefore unsurprising that spin-coated thin films display vivid texture, ranging from the crystallographic to the mesoscopic length scales. These morphological and structural heterogeneities within thin films have been linked to avoidable variations in photovoltaic metrics^[3]. Finer control over photovoltaic parameters necessitates a better understanding and control over how the non-trivial perovskite crystallization is guided, and what impact do different film transformation methodologies have on the transient and emergent functional properties of the material. Notwithstanding the impact of morphological heterogeneity on photovoltaic performances, the chemical and physical history of the material treatment protocols is reflected within the photovoltaic response of the device. The direct consequence of the above factors gives rise to large variations in photovoltaic properties and diminishes the predictability of the material behavior. It has remained a challenge, until now, to correlate the impact of material structure to its functional properties, due to the absence of appropriate measurement setups that allow the simultaneous resolution of structural and electronic changes during spin-coating, especially when crystallization is enforced through nucleation by using orthogonal solvents.

In the current study, the premier case of the in-situ evolution of spin-coated hybrid perovskites thin films is presented through multimodal techniques, made possible by a custom-designed measurement chamber. Grazing incidence wide angle x-ray scattering^[4,5] is utilized to probe the evolving crystalline structure of the spin-coating thin film. Optoelectronic response of the dynamically evolving material is measured through photoluminescence. The film, after spin-coating is annealed and phase transformation of the intermediate state is regulated by temperature, which is controlled through a previously calibrated, spot-regulated pyrometer. Local changes in temperature, emissivity of the resultant thin film are characterized by IR radiographic imaging. The simultaneous, highly time-resolved measurements enable unraveling many mysteries and provide previously inaccessible insights about of the exciting analogue of organic-inorganic lead halide perovskite materials, as well as the kinematic behavior of solution processed soft materials.

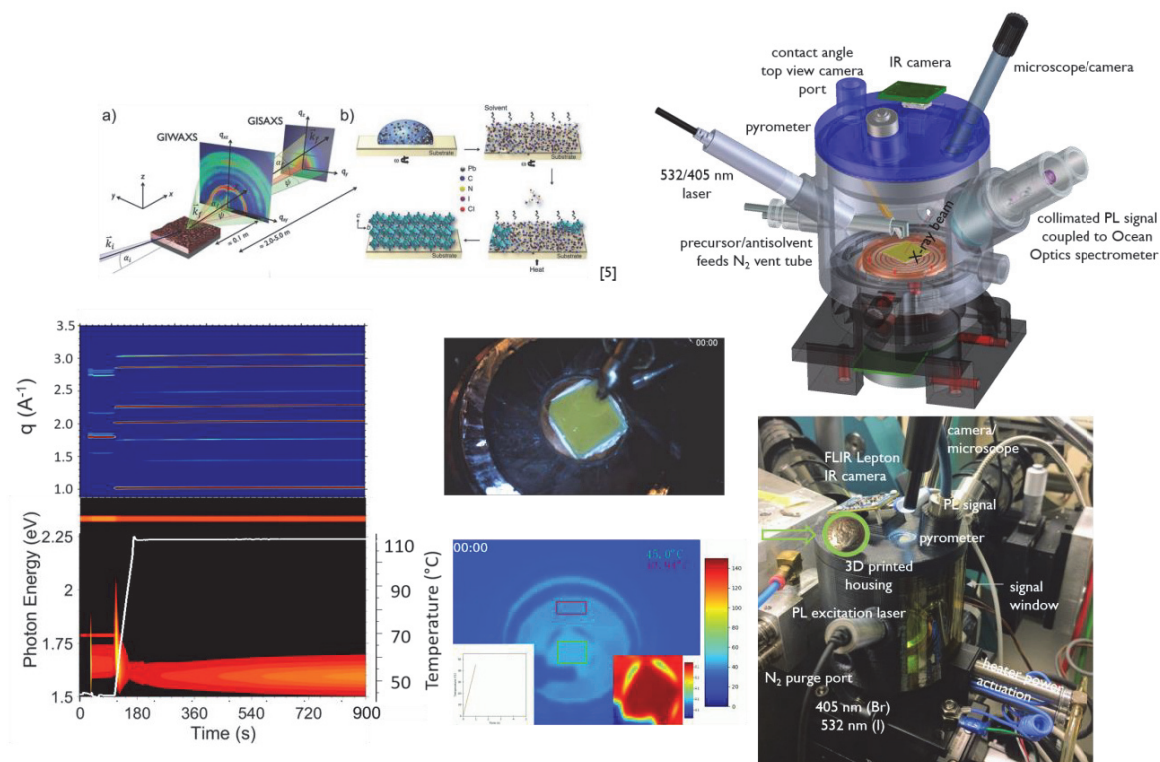


Fig. 1 Engineering drawings and images of the designed measurement chamber deployed at the ALS 12.3.2 microdiffraction beamline and exemplary data output, for studying solution processed optoelectronic / photovoltaic thin films

References

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