CPP 24: Hybrid and Perovskite Photovoltaics III

Time: Wednesday 9:30–11:15 Location: H38

CPP 24.1 Wed 9:30 H38

Tailored perovskite crystallization by passivation molecule engineering for efficient light-emitting diodes — •Jungui Zhou[1,2], Min Zhu[1], Yufeng Zhai[2], Shouzheng Chen[2,4,5], Benedikt Sochor[2], Sarathlal Koyiloth Vayalil[2], Lei Cai[3], Man-Keung Fung[1,6], Peter Müller-Buschbaum[5], and Stephan V. Roth[2,7] — [1]Soochow University, Suzhou, China — [2]DESY, Hamburg, Germany — [3]Shandong normal university, Shandong, China — [4]FRM II, Garching, Germany — [5]TUM School of Natural Sciences, Garching, Germany — [6]MUST, Macau, China — [7]KTH Royal Institute of Technology, Stockholm, Sweden

Metal halide perovskite light-emitting diodes (PeLEDs) are regarded as alternative candidates for next-generation display technologies.Various additives have been widely used in perovskite precursor solutions, aiming to improve the as-obtained perovskite film quality through passivating defects and controlling the crystallinity. Although the defect passivation of additives has been intensively investigated, a deep understanding of how additives influence the crystallization process of perovskites during the spin-coating and annealing processes is still lacking. Here, by combining In-situ photoluminescence (PL) and grazing-incidence wide/small-angle X-ray scattering (GIWAXS/GISAXS) techniques, a systematic study of the perovskite film-formation process, perovskite structure, and inner morphology of CsPbBr3 perovskite films modified by various additives is conducted, revealing the influence of additives on the formation of high-quality perovskite films and efficient PeLEDs.

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Optical In-Situ Methods as Process Optimization Toolbox — •Lennart Reb — Helmholtz-Zentrum Berlin, Hahn-Meitner-Platz 1, 14109, Berlin

In the field of perovskite photovoltaics, precursor optimization is often guided by the visual inspection of film quality by experienced researchers. However, combined optical in-situ techniques can help shift toward rational, evidence-based process design. In-situ characterization methods are frequently used for studying the reaction kinetics during processing: Optical techniques to monitor solvent evaporation, such as spectral absorption, reveal changes in halidoplumbate spectral signatures that correlate with the increasing density of polynuclear plumbate species. Photoluminescence (PL) measurements further elucidate early perovskite nucleation during quenching processes and crystal growth. Structural insights from grazing-incidence X-ray scattering (GIWAXS) track intermediate phases, the formation of perovskite structures, and the evolution of secondary phases. This work briefly reviews the state-of-the-art multimodal in-situ characterization techniques and introduces the multimodal μ slot-die coater, a small-scale platform integrating UV-vis, PL, imaging, and GIWAXS capabilities. Initial results of combined measurements demonstrate its power for studying perovskite film formation during scalable processing, offering insights into reaction kinetics and showcasing the capabilities of in-situ imaging techniques in scalable perovskite deposition. For example, in-situ PL measurements show the changes of PL signal over time upon the inclusion of chloride-based additives.

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Metal halide perovskite solar cells under space like temperature conditions — •Simon Alexander Wegener[1], Altantulga Buyan-Arivjikh[1], Kun Sun[1], Zerui Li[1], Xiongzhuo Jiang[1], Matthias Schwarzkopf[2], and Peter Müller-Buschbaum[1] — [1]TUM School of Natural Sciences, Chair for Functional Materials, 85748 Garching, Germany — [2]Deutsches Elektronen-Synchrotron (DESY), 22607 Hamburg, Germany

Perovskite solar cells hold great promise for space applications due to their exceptional properties, including high power-to-weight ratios and efficiencies comparable to silicon cells. Their solution processability lowers both manufacturing and launch costs, presenting a cost-effective alternative to gallium arsenide cells. However, their viability in space is challenged by harsh conditions such as high vacuum, extreme temperatures, and radiation. This study investigates the impact of extreme temperature fluctuations in low Earth orbit, ranging from -100*C to +100*C, on perovskite solar cell performance. Operando GIWAXS measurements enable real-time analysis of the crystal structure under simultaneous illumination and thermal cycling. Measurements of I-V curves and optical absorption spectra further assess electrical and optical properties. Results reveal temperature-dependent efficiency variations and degradation influenced more by device layers and interfaces than the active layer itself. Understanding the mechanical, optical, and electrical behavior of the entire cell assembly under such conditions is key to optimizing durability and performance.

CPP 24.4 Wed 10:15 H38

Simulation of the impact of processing conditions for solution-processed thick perovskite layers — •Martin Majewski[1], Shudi Qiu[2], Larry Lüer[2], Vincent M. Le Corre[2], Tian Du[2], Olivier J.J. Ronsin[1], Christoph J. Brabec[2], Hans-Joachim Egelhaaf[2], and Jens Hartin[1] — [1]Helmholtz Institue Erlange-Nuernberg, Cauerstraße 1, 91058 Erlangen, Germany — [2]Institute of Materials for Electronics and Energy Technology (i-MEET), Friedrich-Alexander Universitaet Erlangen Nuernberg

Fabricating thick (1000 nm) solution-processed perovskite layers is expected to increase the efficiency of carbon-contact-based solar cells compared to thinner (500 nm) films. However, increasing only the deposited layer thickness often results in buried voids inside the dry film. Recently, we have developed a theoretical framework based on Phase Field simulations. With the help of the simulations, it is possible to explain why voids form in the film. The crystals nucleate at random spots inside the liquid film. The movement of the condensed-vapor interface, due to evaporation, leads to an agglomeration of the crystals at the film surface. The crystals block further evaporation and the remaining solvent is the origin of the buried voids inside the dry film. We explain how adding seeds on the substrate before coating the thick film can prevent this. In this case, processing conditions have to be modified compared to standard operating procedures for thin films. The theoretical expectations can be verified experimentally, leading to a performance improvement of the devices.

CPP 24.5 Wed 10:30 H38

First-principles modelling of hybrid perovskites — •Udo Schwingenschlögl and Aleksandra Oranskaia — King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Saudi Arabia

The photoconversion efficiency record of silicon-perovskite solar cells exceeds 30% owing to hybrid perovskites with organic cations that stabilize the perovskite by non-covalent bonding. To address the electronic properties and stability issue from the perspectives of the bulk crystal phases, point defects, and surfaces and interfaces (requiring large simulation cells) the computational methods must be chosen carefully: (1) For the structural relaxation an exchange-correlation functional is required that adequately describes materials rich in NH$\cdots$I bonding (between organic cations and I) and I$\cdots$I bonding (between PbI_6 octahedra or between PbI_6 octahedra and I-related defects). (2) For the electronic structure calculation an exchange-correlation functional is required that adequately describes the spin-orbit coupling of the Pb and I electrons. Comparing the PBE, PBE-TS, PBE-D3, PBEsol, vdW-DF2, and rVV10 functionals for relaxing FAI, $C_4N_2H_{12}(I_3)_2$, $C_6H_7NI(I_3)$, I_2, In, $Cs(I_3)$, $Cs_2(I_3)_2(I_2)$, and PbI_2 crystals, we show that the rVV10 functional provides the most balanced prediction for the types of non-covalent bonding relevant for hybrid perovskites. We also discuss problems related to the HSE06 functional and show that the PBE functional with a Hubbard correction for the Pb 6p and I 5p orbitals (together with fully relativistic pseudopotentials) provides promising results.

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An effective Population balance model for evaporation-driven precursor-mediated crystallization — •Kai Segadlo, Olivier Ronsin, and Jens Harting — Helmholtz Institute Erlangen-Nürnberg for Renewable Energy (IET-2), Erlangen, Germany

Although tremendous progress has been made in recent years in Experimental and Material Design for solution-processed photoactive Perovskite thin films, this progress has not likewise been followed by a corresponding improvement in the theoretical understanding of these systems. In particular, the high computational demands arising from the plethora of potentially relevant thermodynamic processes, such as

evaporation, crystallization, chemical reactions, demixing, and advection, as well as the high costs of in situ multichannel screening experiments, lead to uncertainties about the underlying physical mechanisms, which in practice often facilitate the use of narrow empirical models. As a first step towards theoretical coherence, we combine empirical mass transport models into a single population balance model accounting for evaporation, crystallization, and chemical reactions. We validate the drying subpart against drying curves, and the crystallization subpart against crystallinity curves from Ultraviolet Imaging Spectroscopy, and Phase field simulations, respectively. The model allows us to gauge the relevances of the processes during precursor-mediated methylammonium lead iodide crystallization measured with in situ Grazing Incidence Wide Angle X-ray scattering and to shed light on the experimentally highly relevant but hard-to-access coupling between evaporation and crystallization.

CPP 24.7 Wed 11:00 H38

Modeling and Analysis of Spectral and Thermal Effects in 2-Terminal Perovskite-CIGSe Tandem Solar Cell Configurations — •Yokozuna Schirmer[1], Nicolas Otto[1], Guillermo Farias Basulto[2], Rutger Schlatmann[1,2], Bert Stegemann[1], and Christof Schultz[1] — [1]HTW Berlin - University of Applied Sciences, Wilhelminenhofstr. 75a, D-12459 Berlin, Germany — [2]PVcomB / Helmholtz-Zentrum Berlin für Materialien und Energie, Schwarzschildstr.3, D-12489 Berlin, Germany

Tandem solar cells are being developed to exceed the efficiency limits of single-junction cells. Typically, they are designed in a 2-terminal (2T) current-matched configuration, which can result in a reduced energy yield due to current mismatch caused by spectral variations throughout the day. To overcome this limitation, tandem solar cells can be configured in a 2T voltage-matched setup, which could mitigate the impact of spectral sensitivity on performance. The aim of this work is the development of a computational model to analyse the energy yield of each configuration. This model combines temperature-dependent electrical cell parameters with spectrally resolved real-world outdoor data to simulate the operational behaviour and to evaluate the advantages of each interconnection approach. The model is validated by measured I-V cell data from single- and multi-junction cells. Moreover, a comprehensive analysis of the calculated results provides valuable insights into the performance trade-offs under real-world conditions, enabling the identification of the optimum design for specific applications.